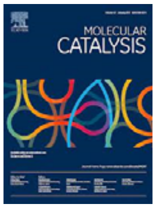
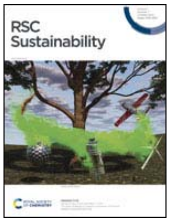
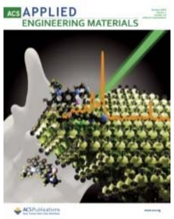
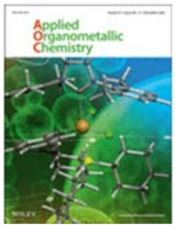
List of Patents & Publications & List of Conference Symposia/Seminars/ Workshops

List of Patents & Publications (Related to Thesis)

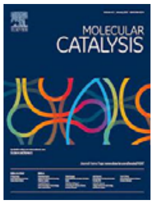
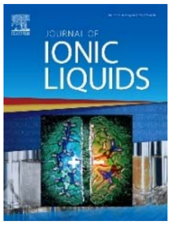
1	INTELLECTUAL PROPERTY **INDIA** PATENTS | DESIGNS | TRADE MARKS GEOGRAPHICAL INDICATIONS	METHOD FOR SYNTHESIS OF *L*-PROLINE GRAFTED ON SILICA SUPPORTED CATALYST AND ITS USE IN ASYMMETRIC ALDOL REACTION **Pratikkumar Lakhani**, Chetan K. Modi **Application number:** 202121048846 **Publication date:** 07/01/2022
2	INTELLECTUAL PROPERTY **INDIA** PATENTS | DESIGNS | TRADE MARKS GEOGRAPHICAL INDICATIONS	BINOL-RU GRAFTED ONTO A SILICA SUPPORTED CATALYST: METHOD OF SYNTHESIS AND ITS USE IN ASYMMETRIC HYDROGENATION REACTION **Pratikkumar Lakhani**, Chetan K. Modi **Application number:** 202221051932 **Publication date:** 07/10/2022
3	INTELLECTUAL PROPERTY **INDIA** PATENTS | DESIGNS | TRADE MARKS GEOGRAPHICAL INDICATIONS	A TAILORED MONTMORILLONITE-SILICA-GRAPHENE OXIDE-BASED COMPOSITE FOR ENHANCING CHIRAL CATALYTIC APPLICATION **Pratikkumar Lakhani**, Chetan K. Modi **Application number:** 202321041553 **Publication date:** 01/09/2023
4		Spick-and-span protocol for designing of silica-supported enantioselective organocatalyst for the asymmetric aldol reaction **Pratikkumar Lakhani**, Chetan K. Modis ***Mol. Catal.*** *2022, 525, 112359* **DOI: 10.1016/j.mcat.2022.112359**
5		Asymmetric Hydrogenation using Covalently Immobilized Ru-BINOL-AP@MSNs Catalyst **Pratikkumar Lakhani**, Chetan K. Modi ***New J. Chem.*** *2023, 47, 8767-8775* **DOI: 10.1039/D3NJ00495C**

6		DFT stimulation and experimental insights of chiral Cu(II)-salen scaffold within the pocket of MWW-zeolite and its catalytic study **Pratikkumar Lakhani**, Darshil Chodvadiya, Prafulla K. Jha, Vivek Kumar Gupta, Damian Trzybiński, Krzysztof Wozniak, Krzysztof Kurzydłowski, U. K. Goutam, Himanshu Srivastava, Chetan K. Modi ***Phys. Chem. Chem. Phys.*** *2023, 25, 14374-14386* **DOI: 10.1039/D3CP00857F**
7		Sustainable approach for the synthesis of Chiral β-aminoketones using Encapsulated Chiral Zn(II)-Salen complex **Pratikkumar Lakhani**, Sanjeev Kane, Himanshu Srivastava, U. K. Goutam, Chetan K. Modi ***RSC Sustain.*** *2023, 1, 1773-1782* **DOI: 10.1039/D3SU00210A**
8		Enhanced Thermal Stability and Catalytic Performance of Montmorillonite-Silica-Graphene Oxide Composite Incorporating with Chiral Thiourea for the Strecker Reaction **Pratikkumar Lakhani**, Chetan K. Modi ***ACS Appl. Mater. Interfaces,*** *2023* (Under communication)

Publication (Non-related to Thesis)

1		Efficient Visible Light Active Photocatalyst: Magnesium Oxide Doped Graphitic Carbon Nitride for Knoevenagel Condensation Reaction Dhavalkumar Bhanderi, **Pratikkumar Lakhani,** Ashita Sharma, Saurabh S. Soni, Sanjeev Kane, U.K. Goutam, Chetan Modi ***ACS Appl. Eng. Mater.*** *2023, 1, 10, 2752–2764* **DOI: 10.1021/acsaenm.3c00463**
2		Harnessing Bimetallic Nanoparticles on Ionic Liquid Functionalized Silica for Enhanced Catalytic Performance Rithik Parmar, **Pratikkumar Lakhani,** Dhavalkumar Bhanderi, Sanjeev Kane, U.K. Goutam, Chetan Modi ***Appl. Organomet. Chem.,*** *2023* (Under communication)

List of Review Article and Book Chapter

Review Article		
1		Shaping Enantiochemistry: Recent Advances in Enantioselective Reactions via Heterogeneous Chiral Catalysis **Pratikkumar Lakhani**, Chetan K. Modi* ***Mol. Catal.*** *2023, 548, 113429* **DOI: 10.1016/j.mcat.2023.113429**
2		Advances in Green Synthesis and Sustainable Processing via Heterogeneous Catalysis: A Comprehensive Review **Pratikkumar Lakhani**, Dhavalkumar Bhanderi, Chetan K. Modi* ***J. Chem. Eng.,*** *2023* (Under communication)
3		Graphitic Carbon Nitrate (g-C3N4) as an Emerging Photocatalyst for Sustainable Environmental Applications: A Comprehensive Review Dhavalkumar Bhanderi, **Pratikkumar Lakhani**, Chetan K. Modi* ***RSC Sustain.,*** *2023* (Under communication)
4		Silica-Supported Ionic Liquids as Versatile Catalysts: Synthesis, Applications, and Recent Advances **Pratikkumar Lakhani**, Dhavalkumar Bhanderi, Chetan K. Modi* ***Journal of Ionic Liquids,*** *2023* (Under communication)
Book Chapter:		
1	SPRINGER NATURE	Photocatalytic Water Splitting **Pratikkumar Lakhani**, Komal Trivedi, Chetan K. Modi **Book Title:** Graphene-based Photocatalysts: From fundamentals to applications **Publisher**: Springer Nature (Under communication)

Work presented in Conferences/ Seminars/ Workshops

1	CHEMICAL RESEARCH SOCIETY OF INDIA ESTABLISHED 1999 Indian Institute of Technology Guwahati	Economical and sustainable approach for the synthesis of supported L-proline catalyst for asymmetric aldol reaction **Pratikkumar Lakhani**, Chetan K. Modi* **National Conference** of Chemical Research Society of India 28th National Symposium in Chemistry (CRSI NSC-28), 25-27th March 2022 at IIT Guwahati. **Presented Session: POSTER**
2	ROYAL SOCIETY OF CHEMISTRY Commonwealth Chemistry Federation of Chemical Sciences Societies	Strategic Design of Silica-supported Enantioselective Organocatalyst for Asymmetric Aldol Reaction: An Economical and Sustainable Approach **Pratikkumar Lakhani**, Chetan K. Modi* **International conference** of 3rd Commonwealth Chemistry Posters. Royal Society of Chemistry, 28-29th September 2022 **Presented Session: POSTER**
3	Interdisciplinary Symposium Materials Chemistry ISMC SOCIETY FOR MATERIALS CHEMISTRY	Catalytic Behaviour of Alkali Treated Meso- and Microporous Silica-based Materials **Pratikkumar Lakhani**, Niraj V. Rana, Chetan K. Modi* **National Conference** of DAE-BRNS 9th Interdisciplinary Symposium on Materials Chemistry (ISMC-2022) Chemistry Division, Bhabha Atomic Research Centre, and Society for Materials Chemistry, 7-10th December 2022 at Mumbai. **Presented Session: POSTER**
4	CHEMICAL RESEARCH SOCIETY OF INDIA ESTABLISHED 1999 ज.ने.वि. JNU	Development of BINOL-Ru Catalyst Covalently Immobilized on MSNs and Their Application in Asymmetric Hydrogenation **Pratikkumar Lakhani**, Chetan K. Modi* **National conference** of Chemical Research Society of India 30th National Symposium in Chemistry (CRSI NSC-30), 2-5th February 2023 at JNU New Delhi. **Presented Session: POSTER**
5	SRM INSTITUTE OF SCIENCE & TECHNOLOGY (Deemed to be University u/s 3 of UGC Act, 1956)	Biologically Potent 3,4-dihydropyrimidine-2-(1*H*)-one Derivatives Synthesized Through Chiral Cu(II)-salen Encapsulated in MWW **Pratikkumar Lakhani**, Chetan K. Modi* **International conference** on 7th International Conference on Nanoscience and Nanotechnology – ICONN 2023 (Virtual Conference) Organised by SRM Institute of Science & Technology, Kattankulathur, Tamil Nadu. **Presented Session: POSTER**

Participated:		
6		1st International Conference on Emerging Porous Materials (ePorMat-2021) A Virtual Conference held on 29-30th July, 2021 NISER Bhubaneswar in association with VIT Vellore and IIT Jammu, India.
7		Synergistic Training Program Utilizing the Scientific and Technological Infrastructure (STUTI-21), A One-Week Training Program on R&D Equipment on the theme “ADVANCES IN CHARACTERIZATION OF MATERIALS" on 12-18th September 2022 at Applied Physics Department, The Maharaja Sayajirao University of Baroda, Vadodara, Gujarat.
8		Synergistic Training Program Utilizing the Scientific and Technological Infrastructure (STUTI-21), A One-Week Training Program on R&D Equipment on the theme “Modern Spectroscopic, Thermal and Microscopic Techniques" on 21-27th September 2022 at Defence Institute of Advanced Technology, Pune, MH.
9		STEM Early Researchers by a Ph.D. student of Inspiring India in Research, Innovation, and STEM Education (iRISE) on the theme **“IP and Knowledge Management”** on 28th September 2022 at IISER Pune.
10		Training Program for Developing Skills on **“Advancing Knowledge from Quantum mechanical Perspectives in materials science”** Attended one-day workshop organised on April 15th,2023 at Department of Physics, The Maharaja Sayajirao University of Baroda, Vadodara.

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Editor's choice paper

Spick-and-span protocol for designing of silica-supported enantioselective organocatalyst for the asymmetric aldol reaction

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ABSTRACT

In the present work, we report a ground-breaking strategy in the development of spick-and-span protocol for the synthesis of L-proline chiral scaffold tethered onto the silica matrix i.e., L-proline-(3°amine)-*f*-SiO_2 via the reaction between L- proline methyl ester and N-methyl aminopropyl silica with absolutely no use of protecting/ deprotecting groups. The as-synthesized catalyst was well corroborated through various physicochemical techniques such as ^{13}C CP MAS NMR, X-ray diffraction (XRD), HRTEM, N_2 adsorption−desorption isotherms, elemental analysis and FT-IR spectral studies. Emphatically, L-proline-(3°amine)-*f*-SiO_2 significantly promoted an asymmetric aldol reaction under ambient conditions with no use of redundant organic solvent, bestowing unprecedented activity with 100% conversion and (ee) $>$ 99% enantioselectivity of S-isomer. Moreover, the as-synthesized catalyst was effortlessly recycled seven consecutive times with absolutely zero loss of activity as well.

Introduction

Enantiomerically active compounds are highly precious scaffolds and have garnered great attention in recent years. These compounds have been found to be active in various ranges of research fields [1], especially in the formulation of pharma products. Many contemporary medicinal drugs are chiral and require high levels of enantiopurity in clinical testing as demanded by governing bodies [2]. Therefore, there has been ancient attention to synthetic approaches providing non-racemic products and with current speedy growth in the catalytic asymmetric synthetic procedures.

For various asymmetric transformations especially in pharmaceuticals, a range of homogeneous catalysts has been used in the past decades [3]. Most of the preparative techniques reported so far are based on homogeneous modes that need excess loading of catalyst and strenuous purification techniques. On the other hand, enhancing the catalytic potency and selectivity by dint of support synergism in heterogeneous catalysis is always been an eye-catching approach, in particular, colossal exertions have been devoted towards the grafting of chiral organo-scaffolds on the solid supports for improving their catalytic competence and also to facilitate the recycling and reuse strategy of the catalyst. Despite the truth, the fairy turned out not to be real when it deals with asymmetric reactions [4,5].

The foremost evidence of proline grafted on a solid support for asymmetric aldol synthesis dates back to the year 2003 [6]. L-proline has been in limelight on account of its natural abundance as well as a metal-free substrate which signifies important consideration in terms of economy in pharmaceuticals. Benaglia and co-workers [4] have grafted L-proline onto a poly(ethylene glycol) (PEG) scaffold [7–9]. Polystyrene (beads) [10,11], Merrifield resin[12], ionic liquids [13–16], dendrimers [17] and multi-wall carbon nanotubes (MWCNTs) [18] are some other prevalent supports for L-proline. It always remains a confront particularly when mesoporous silica with flexible mesoporous surfaces, furnishes the best practicable solutions as support [19–26]. Based on the fascinating mass transfer reaction, higher surface area and the ease competence of grafting chiral organic scaffolds, mesoporous silica has been used as effectual support for L-proline and endures for alluring catalytic properties [27–29].

To date, most of the reported methods for the synthesis of silica-supported L-proline catalysts involve multistep and somewhat complex syntheses, involving protecting/ deprotecting steps i.e. protections of the secondary amine of L-proline through t-butyloxycarbonyl (Boc) and carboxylic acid via esterification[30]. In contrast to this, E. Kristensen et al. [31] have reported polymer-supported proline as an organocatalyst utilizing unprotected proline, avoiding the protection/deprotection steps. In line with this, A. A. Elmekawy et al. [32] and Z. An et al. [33] have developed proline immobilized onto silica supports by reacting trans-4-hydroxy-L-proline with chloropropyl tethers with no

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ROYAL SOCIETY OF CHEMISTRY

PAPER

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Asymmetric hydrogenation using a covalently immobilized Ru-BINOL-AP@MSNs catalyst†

Pratikkumar Lakhani and Chetan K. Modi*

The present work describes an unprecedented blueprint for the chiral (*S*)-1,1′-Bi-2-naphthol ligand (BINOL) immobilized on amine functionalized mesoporous silica nanoparticles (MSNs) *via* a linker, which is then converted into a ruthenium complex, *i.e.*, Ru-BINOL-AP@MSNs (where AP = (3-aminopropyl)trimethoxysilane), without the use of deprotecting or protecting groups. Using a variety of techniques, such as FTIR, N_2 adsorption–desorption isotherms, solid-state ^{13}C CP MAS NMR, powder XRD, FESEM, HRTEM, XPS, and thermogravimetric analysis, the as-synthesized catalyst was persuasively verified. Asymmetric transformations of enantiomerically enriched chiral alcohols can be achieved with the aforesaid active catalytic precursors, making the proposed method even more appealing. During the asymmetric hydrogenation reaction, the Ru-BINOL-AP@MSNs catalyst proved to be catalytically competent, leading to an 84% conversion with an enantioselectivity >90% of the *R*-isomer. Furthermore, the synthesized catalyst was capable of being recycled five consecutive times with only a minute loss of its effectiveness, thereby reducing solvent waste and precious metal or ligand losses.

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1. Introduction

Asymmetric reduction of unsaturated compounds presents an excellent opportunity for introducing stereogenic elements along with new functionalities into the structure of organic compounds. As a result, this process has become one of the most popular methods of asymmetric synthesis, and it has been used to prepare a variety of organic compounds of biological interest. It is notable that, among the methodologies currently available for this purpose, hydrogen-transfer reduction occupies the second prominent position, followed by asymmetric hydrogenation using molecular hydrogen.[1,2] Despite having excellent selectivities and yields, most of the literature-reported methods require a stoichiometric amount of sensitive reagents or the utilization of unsafe hydrogen gas at elevated pressure.[3] Due to availability of hydrogen donors (2-propanol), as well as the fact that it is cost effective, convenient, and need not rely on complicated experimental setups such as browbeat reactors, the hydrogen-transfer reduction reaction is gaining popularity.[4] On the other hand, homogeneous metal catalysts (Ru and Ir complexes) are frequently used for these chemical reactions.[5,6] Although a homogeneous catalyst can generally be recovered and its products purified using methods such as purification, extraction or centrifugation, complete removal is difficult to achieve even with these methods.[7] In addition to recycling catalysts, catalysts should also be removed from the reaction mixtures at the level of parts per million. The reason these workups are less suitable for industrial applications is that they are time-consuming, tedious, as well as energy-consuming.

The chiral binaphthyl plays an important role as an auxiliary chiral component.[8] There are various enantioselective catalytic systems based on BINOLs and BINAPs developed by Noyori *et al.*[9] The BINOL-based scaffold has been immobilized by grafting it to organic and inorganic supports. However, we have been researching the immobilization of chiral BINOL species using a contemporary approach. Furthermore, ruthenium metal hydrides produced by asymmetric transfer hydrogenation (ATH) are highly susceptible to air, making their isolation and reusability extremely challenging. There have been a number of heterogeneous variants developed to circumvent these problems, including polymers,[10,11] carbon nanotubes,[12] ionic liquids,[13–15] dendrimers,[16–18] inorganic materials (like Fe_3O_4 nanoparticles),[19,20] or organic/inorganic materials.[7,12] Most of these systems require high temperatures, extended reaction times, an uplifted catalyst loading, excess base, and a time-consuming recovery process, like filtration or centrifugation. The employment of both chiral auxiliaries and transition metal catalysts is often rather expensive, which is why silica supports have been used to immobilize enantioselective catalyst systems to ensure that catalyst separation and recycling is facilitated.[21–23] The use of binaphthyl's chirality in silica

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† Electronic supplementary information (ESI) available. See DOI: https://doi.org/10.1039/d3nj00495c

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DFT stimulation and experimental insights of chiral Cu(II)–salen scaffold within the pocket of MWW-zeolite and its catalytic study†

Pratikkumar Lakhani,[a] Darshil Chodvadiya,[b] Prafulla K. Jha,[b] Vivek Kumar Gupta,[c] Damian Trzybiński,[d] Krzysztof Wozniak,[d] Krzysztof Kurzydłowski,[e] U. K. Goutam,[f] Himanshu Srivastava[g] and Chetan K. Modi *[a]

A Cu(II)–salen complex encapsulated in MWW-framework as an efficient chiral organocatalyst was developed for the synthesis of 3,4-dihydropyrimidin-2-(1*H*)-one (DHPMs) derivatives *via* an asymmetric pathway. In order to confirm its structural properties, single-crystal X-ray diffraction, powder XRD, BET, XPS, FE-SEM, EDX, UV-Vis, and FTIR spectra were used. Using computer-assisted DFT calculations, the Cu(II)–salen complex has been fine-tuned to fit into the pocket of the porous MWW support while keeping its chirality. This organocatalyst was shown to be a potent catalyst for the formation of the desired DHPMs product under short reaction times. Furthermore, this green protocol allows rapid and simple isolation of active MWW-trapped Cu(II)–salen scaffolds and its reusability in at least five consecutive runs without losing much of its activity.

1. Introduction

There have been numerous natural and synthetic bioactive substances developed using asymmetric synthetic techniques, including a multitude of total syntheses.[1] Multicomponent reactions (MCRs) can be used to synthesize organic compounds in a single step with a high atom economy. For instance, natural products have been synthesized using MCRs.[2] Despite the fact that chiral induction in MCRs is possible, it is not simple, and the selectivities that can be achieved aren't always ideal.[3]

As a method for preparing DHPMs, the multicomponent for instance, Biginelli-like reactions concerning aldehydes, urea and/or thiourea, and enolizable carbonyls have proven particularly effective. DHPMs can be used to treat infections and parasites like malaria,[4,5] and they can also be used to reduce high blood pressure.[6] Despite the Biginelli reaction's long history of discovery, asymmetric routes have received relatively little attention. It wasn't until 2003 when the reaction of urea, benzaldehyde, and methyl acetoacetate with cerium(III) chloride utilizing a chiral amide was first described; this reaction was facilitated by Wang J. H. and teammates.[7] Despite this, the enantiomeric excess (ee) that was achieved was just 40%. The asymmetric Biginelli reaction, which uses a chiral ytterbium catalyst to achieve high enantioselectivities (>99% ee), was next described by Huarig Y. *et al.*[8] Chen X. H. and co-workers later described the foremost organocatalytic Biginelli reaction, which provided DHPMs with ee values as high as 97% by employing chiral phosphoric acids derived from 1,1′-bi-2-naphthol (BINOL) as catalysts.[9] However, there are drawbacks, such as the need for extremely hazardous organic solvents, a significant amount of catalyst, severe reaction conditions, and a lot of time and effort spent on separating the target product. Therefore, there is a need for the creation of straightforward Biginelli techniques that are gentle and environment caring, and recently, some intriguing environmentally friendly

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† Electronic supplementary information (ESI) available. CCDC 2181683. For ESI and crystallographic data in CIF or other electronic format see DOI: https://doi.org/10.1039/d3cp00857f

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Sustainable approach for the synthesis of chiral β-aminoketones using an encapsulated chiral Zn(II)–salen complex†

Pratikkumar Lakhani,[a] Sanjeev Kane,[b] Himanshu Srivastava,[b] U. K. Goutam[c] and Chetan K. Modi *[a]

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To enable sustainable chemical transformations, it is imperative to adopt ecofriendly strategies aligned with economic growth and environmental preservation. We present a more environmentally conscious method for synthesizing Zn(II)–salen ligand encapsulated within an MWW host as a heterogeneous chiral catalyst, denoted as Zn(II)–salen@MWW. Various techniques, including FTIR, FESEM, EDX, XRD, BET, and XPS were used to confirm the successful chiral Zn(II) ligand encapsulation. Utilizing an uncomplicated ultrasonic approach, the synthesized catalyst efficiently produces chiral β-amino carbonyl compounds at room temperature under solvent-free conditions. The catalytic process takes 120 minutes, yielding an impressive 94% with selectivity >94%. This protocol offers multiple benefits, including an environmentally friendly catalyst, simple setup, easy separation, and the capability to reuse the chiral Zn(II)–salen@MWW catalyst for up to five runs. Substrate tests involving aldehydes, ketones, and anilines exhibit yields ranging from 96% to 80% and selectivity from 96% to 83%. The process holds significant potential for academia and industry.

Sustainability spotlight statement

The pursuit of sustainable chemical transformations necessitates the integration of eco-friendly approaches, economic growth, and environmental conservation. In line with this objective, a remarkable breakthrough has been made in the development of a greener decorum for synthesizing the present catalyst. The present heterogeneous chiral catalyst exhibits immense potential for promoting sustainable practices in the field of asymmetric synthesis. The utilization of an unpretentious ultrasonic approach allows the synthesized catalyst to efficiently generate chiral β-amino carbonyl compounds under solvent-free conditions at room temperature. This significantly contributes to the reduction of both energy consumption and the generation of toxic byproducts. Another noteworthy feature is the effortless separation of the catalyst from the reaction mixture, facilitating its recovery and reusability. The ability to reprocess the catalyst for up to five runs not only minimizes waste generation but also optimizes resource utilization. This further reinforces the sustainability aspect of the process, as it promotes the efficient use of materials and reduces the demand for fresh catalyst production. As efforts towards sustainability continue to gain momentum, this innovative approach is poised to play a crucial role in advancing both academic research and industrial practices.

Introduction

Asymmetric catalysis has become highly advanced in recent years, but there are still challenges to overcome in this area, such as developing environmentally safe methods and using substrates that have been considered unreactive.[1] Ketones, in particular, pose a significant challenge to the current state of asymmetric methodologies.[2] Due to their low reactivity and difficulties in restraining facial stereoselectivity, some enantioselective chiral catalytic C–C bond formation reactions with carbonyl are available, despite being effective procedures for enantioselective reduction of ketones.[3] In contrast, salen–metal complexes may be useful as bifunctional Lewis acid–base catalysts for contemporary catalytic reactions with imperative substrates.[4]

Scientists have embraced Green Chemistry principles over the past few decades, and environmental considerations have become part of chemical processes.[5] A significant part of the active pharmaceutical ingredient (API) manufacturing processes relies on the reaction media, which accounts for up to 80% of the total mass.[6,7] To overcome this issue, the major pharmaceutical companies have developed solvent selection guides for drug synthesis chemical processes. The development of clean technologies that replace hazardous organic solvents with environmentally friendly solvents has become increasingly

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† Electronic supplementary information (ESI) available. See DOI: **https://doi.org/10.1039/d3su00210a**

ACS APPLIED ENGINEERING MATERIALS

Efficient Visible Light Active Photocatalyst: Magnesium Oxide-Doped Graphitic Carbon Nitride for the Knoevenagel Condensation Reaction

Dhavalkumar Bhanderi, Pratikkumar Lakhani, Ashita Sharma, Saurabh S. Soni, and Chetan K. Modi*

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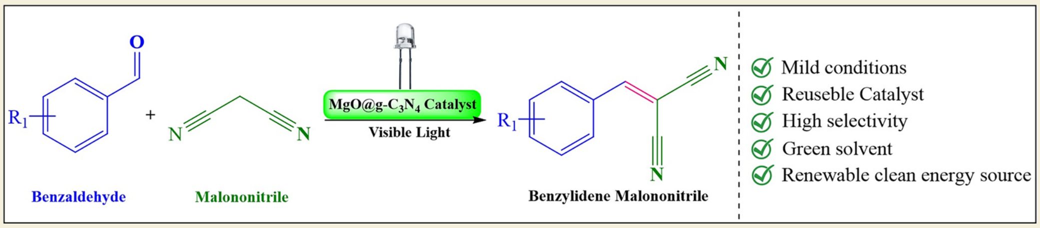

ABSTRACT: In the present work, we have developed visible light active photocatalysts by incorporating magnesium oxide (MgO) as a dopant (with varying loadings, viz., 3, 5, and 10%) onto graphitic carbon nitride (g-C_3N_4) nanosheets prepared via urea and dicyandiamide as precursors to form MgO@g-C_3N_4 catalysts. These photocatalysts showed impressive photocatalytic performance in the Knoevenagel condensation reaction using a low-power visible LED light (12 W), surpassing that of pure g-C_3N_4. In order to characterize photocatalysts, various techniques have been used, such as FTIR, XRD, XPS, TEM, EIS, UV−vis DRS, and thermogravimetric analysis. Based on this study, the MgO@g-C_3N_4 catalyst with 5% MgO loading has shown an outstanding performance with 98.53% conversion and 98.98% selectivity. Furthermore, the catalyst showed minimal drop-in activity after five cycles of recyclability. This study highlights MgO@g-C_3N_4 as a promising and sustainable photoactive material for efficient photocatalytic applications.

KEYWORDS: *graphitic carbon nitride (g-C_3N_4), MgO, photocatalyst, visible light, Knoevenagel condensation*

INTRODUCTION

In recent years, the progress in new photocatalytic materials has attracted significant interest due to their potential across various environmental and energy applications.[1,2] Significantly, graphitic carbon nitride (g-C_3N_4) presents itself as a potential photocatalyst under visible light due to its distinct electronic band arrangement, exceptional durability, and ample elemental composition.[3−8] The semiconductor's adjustable characteristics and notable photoactivity render it an appealing contender for a wide array of photocatalytic applications, including pollutant degradation, water splitting, and organic synthesis.[9−13] The typical synthesis process of g-C_3N_4 involves the thermal polymerization of nitrogen-rich precursors such as urea and dicyandiamide, yielding a layered arrangement featuring interconnected heterocyclic units. Despite its advantageous properties, g-C_3N_4 suffers from certain limitations such as limited visible light absorption and low charge carrier mobility, which can hinder its photocatalytic efficiency. To address these challenges and improve the photocatalytic performance of g-C_3N_4, researchers have investigated different approaches, such as incorporating metal or metal oxide nanoparticles through doping.[14−21] Among these, magnesium oxide (MgO) has shown great promise as an effective dopant thanks to its wide band gap, high thermal stability, and excellent charge-transfer properties. Through the incorporation of MgO as a dopant, it becomes feasible to customize the electronic band configuration of g-C_3N_4. This alteration extends its capacity for absorbing light and enhances the separation and mobility of charge carriers.[22−24]

During the past few years, light-assisted organic synthesis has grown rapidly.[25,26] One of the most fundamental transformations in organic chemistry is the formation of a carbon−carbon bond in Knoevenagel condensation reactions.[27−30] Many classical acid−base or base-catalyzed heterogeneous systems have been reported in this context.[31−36] Despite their advantages, these systems suffer from numerous drawbacks such as high temperatures, longer reaction times, poor selectivity, and low energy efficiency; therefore, they are not practical. Thus, it is imperative to develop heterogeneous photocatalytic systems that are greener,

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Review

Shaping enantiochemistry: Recent advances in enantioselective reactions via heterogeneous chiral catalysis

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ARTICLE INFO

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ABSTRACT

Asymmetric catalysis refers to the process in which less than stoichiometric concentrations of the catalyst are used for synthesis of one enantiomer preferentially over the other. This review article provides a concise summary of the most current applications, methodologies, and approaches to asymmetric catalysis under heterogeneous conditions. The technique allows subsequent use of chiral catalysts without diminishing activity or enantioselectivity. In this review article, numerous examples of industrial and laboratory chemo catalyzed processes will be presented, such as hydrogenation, epoxidation, Aldol, Diels-Alder reactions, diethylzinc addition, and ketone reduction. There is a general phenomenon called the confinement effect on surfaces and pores which can increase or decrease enantioselectivity.

1. Introduction

In light of the continuously growing demand for nonracemic chiral compounds, the introduction of effective procedures for creating enantiomerically enriched products holds significant value for both the industrial and academic sectors [1,2]. In recent decades, catalysis has become an integral part of the Green Chemistry approach, and its ongoing advancements have enabled us to replace outdated stoichiometric techniques with more environmentally friendly ones [3]. Asymmetric catalysis is one of the most all-encompassing and attractive approaches among those used for this purpose in terms of chiral conformity and ecological concerns [4–7]. In organic transformations, catalysts play a remarkable role in the full conversion of reactants into products under benign reaction conditions. As a result of homogeneous catalysts' high reactivity, selectivity, and low loading requirements, chemists rely on them a lot [8]. Despite homogeneous catalysts' benefits, they are prone to contamination with metal product and low catalyst reusability [9,10]. The range of catalytic reactions that's able to be carried out with excellent enantioselectivity and efficiency has been substantially increased over the last several decades because of significant investigation in this field. For the homogenous chiral catalysis of diverse organic reaction, numerous chiral ligands and their Metal coordination compounds have been invented [11,12]. The key issue in this scenario is the availability of recyclable chiral catalysts for industrial applications. In sectors where reactions are carried out in large quantities, homogeneous catalysts present a major economic loss due to the loss of precious metal catalysts that can't easily be recovered. Taking into account the substantial cost of both metals and enantioselective ligands, systems that enable simple, effective reutilization of enantioselective catalysts are highly desirable [13]. Although essential for industrial manufacturing, homogeneous catalytic methods frequently struggle to achieve this. Another major drawback of homogeneous catalytic processes is the effects of metal leaching, which are detrimental to the production of fine chemicals and pharmaceuticals. Such problems can be solved with heterogeneous asymmetric catalysis, which is gaining popularity these days [14]. This type of catalysis employs both enantioselective designed heterogeneous metal catalysts for asymmetric transformation and immobilized homogeneous asymmetric catalysts [15,16]. Catalyst immobilization is the approach of converting a homogeneous catalyst into a heterogeneous catalyst that can be isolated from the reaction medium and, preferably, used repeatedly [17–19].

Nevertheless, this rarely holds true in practical scenarios, and a homogeneous catalyst's immobilization can suffer from a variety of issues. For instance, a supported chiral catalyst may exhibit decreased catalytic properties (or even total inactivation) when correlated to a homogeneous catalyst because the active sites are less accessible in the solid matrix. Other than that, interactions with the support can unexpectedly affect the silhouette of an improved homogeneous catalyst, which regularly leads to a decrease in enantioselectivity. Due to these factors, it is usual preparation to utilize a length of linker enough to attach the

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Prof. T. Punniyamurthy
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Mr. PRATIKKUMAR C. LAKHANI

from The Maharaja Sayajirao University of Baroda, Vadodara has participated in 7-day training program held at Applied Physics Department, Faculty of Technology & Engineering, The Maharaja Sayajirao University of Baroda, Vadodara, Gujarat.

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during 21st -27th September 2022 at DIAT, Pune.

Dr. T. K. Sai
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Pratikkumar Chimanbhai Lakhani

a Ph.D. student of The Maharaja Sayajirao University of Baroda has attended the one-day workshop organised on September 28, 2022, at IISER Pune for STEM Early Career Researchers by Inspiring India in Research, Innovation, and STEM Education (iRISE) in collaboration with CSIR-National Chemical Laboratory on "**IP and Knowledge Management**".

C Harinath

Prof. Harinath Chakrapani

Professor, Dept of Chemistry,
Indian Institute of Science Education and Research Pune
Project Investigator, iRISE

Dr. Nitin Shukla

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(Prof. Prafulla K. Jha)
Head, Department of Physics