

Green and Sustainable Access to Thiazoles and Benzothiazoles: The Promising Role of Some Metal Oxide Nanocatalysts

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Abstract—Thiazole and benzothiazole structural frameworks have long been regarded as privileged heterocyclic frameworks in medicinal chemistry, as they form the basis for a diverse range of commercially available drugs with varying biological activities. Conventional synthesis methods, such as the Hantzsch reaction, have paved the way for synthesizing thiazole and benzothiazole derivatives; however, many of these methods involve toxic reagents, significant amounts of waste, or have unwanted reaction conditions that compromise the principles of green chemistry. This comprehensive review will critically and comprehensively assess the expanding and developing role of metal oxide nanoparticles (MONPs) as efficient and sustainable heterogeneous catalysts for the synthesis of thiazole and benzothiazole derivatives. Metal oxide nanoparticles, including ZnO, CuO, Fe₂O₃, TiO₂, ZrO₂, and Bi₂O₃, have a high surface area, thermal stability, tunable acidity and basicity, and are easily separated and reused, making them ideal for green synthesis. The review emphasizes their successful implementation into one-pot, multicomponent reactions (MCRs), which are atom-economic and convergent. Their key advances are reported, mainly the extraordinary efficiency of ZnO NPs, which reaches 93% in 7.0 minutes, as well as their magnet separability of Fe₂O₃ NPs, making catalyst recovery convenient, and their synergistic effect on nanocomposites like Fe₃O₄@SiO₂.TiO₂. The review concludes that nano catalysis, especially with MONPs, is a strong, green chemistry framework that overcomes the challenges of classical methods in preparing these important heterocycles.

Keywords—Thiazoles, Nanocatalyst, eterocyclic compounds, Sustainable environment, Green Chemistry.

I. INTRODUCTION

The synthesis of several small heterocycles possessing significant biological activity. Heterocycles containing nitrogen [1], sulfur [2], and oxygen [3] heteroatoms form the essential basis for the development of novel biologically active compounds in the pharmaceutical industry. Thiazoles and benzothiazoles, in particular, form a significant portion of the sulfur-containing heterocycles and are an essential scaffold found in several pharmaceutically important compounds and natural products, which possess many different biological activities (antiviral, anticancer, and antimicrobial) [4]. Some of the approved drugs containing the thiazole moiety[1] are shown in Figure 1. Thiazole, as

well as benzothiazole (where a benzene ring is fused), consists of a five-membered ring system containing S and N heteroatoms at sites 1 and 3, respectively.

The medicinal importance of this scaffold has led many chemists to subsequently report a large number of reports on the synthesis of thiazole derivatives [5]. Traditional methods, such as the Hantzsch thiazole synthesis method[6-10], which exploits a reaction mechanism between halo ketones and thioamides, and subsequent modified methods[11], which include reactions of terminal alkynes, sulfonyl azides, and thio esters (Gabriel synthesis) [12], Cook–Heilbron synthesis [13], etc., were first used to prepare thiazoles. While these methods were convenient, they were not safe or environmentally friendly since they produced significant waste, employed toxic chemicals, and required prolonged high temperatures and time [14-17].

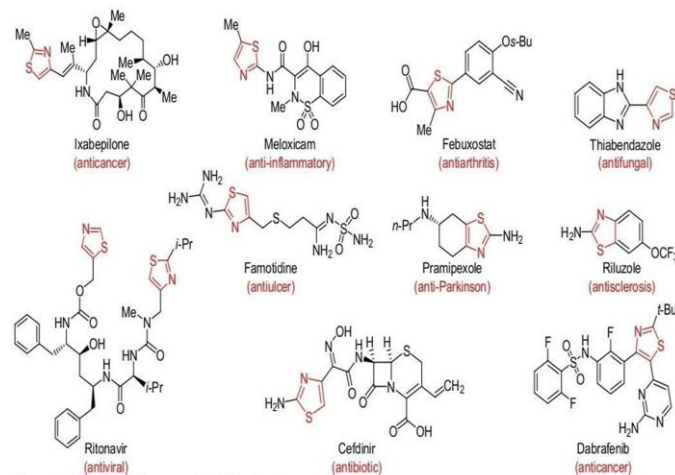


Fig. 1: Some of the approved drugs containing the thiazole moiety

This literature review has served as an impetus for the pursuit of sustainable and effective catalytic systems to support sustainability. In this regard, nanocatalysis has emerged as a powerful new frontier. Nanoparticles, because



they contain both high surface area-to-volume ratios, provide for a high number of active sites, which enables enhanced catalytic activity. Metal oxide nanoparticles (MONPs), specifically, have garnered significant attention, given their properties, which include, but are not limited to: thermal stability, ease of synthesis and functionalization, ability to recycle, and their low toxicity properties. Because of their inherent Lewis/Brønsted acidity or basicity, their tunable nature provides for MONPs to act as ideal heterogeneous catalysts, supporting the cyclocondensation reactions important to the synthesis of thiazole and benzothiazole [18-21]. Therefore, the objective of this literature review is to evaluate the state-of-the-art in the development of metal oxide nanoparticles as catalysts for the efficient and green preparation of thiazole and benzothiazole derivatives, assessing their efficacy, sustainability, and prospects.

Synthesis of thiazole and benzothiazole using metal oxide NPs as a catalyst.

Several metal oxide NPs have been successfully utilized as catalysts for the one-pot synthesis of thiazole and benzothiazole derivatives via multicomponent reactions (MCRs), which are highly convergent and atom-economical.

II. ZINC OXIDE NANOPARTICLES (ZNO NPs)

Zinc oxide (ZnO) nanoparticles (NPs) are one of the most commonly used catalysts for a variety of applications due to their high Lewis acidity, biocompatibility, and ease of preparation [22-24]. We have demonstrated a simple synthetic protocol for the synthesis of 2-aryl-1,3-benzothiazoles using ZnO NPs as a mild and efficient heterogeneous catalyst. The reactions using ZnO NPs were able to proceed at a remarkable rate (< 8 minutes) and yield excellent (> 90%) yields of products. In addition, the catalyst was recyclable and could be reused eight times without losing any significant catalytic activity [25].

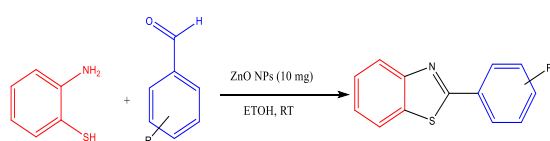


Fig. 2: Synthesis of benzothiazole derivatives using ZnO NPs as a catalyst

Furthermore, a novel and green method for 1,3-thiazole was accomplished via multicomponent reactions between primary amines, isothiocyanates, and alkyl bromides in the absence of solvent when employing nanorod ZnO structures as the catalyst, as shown in Figure 3. In the absence of a catalyst, the reactions did not take place. The catalyst has demonstrated considerable reusability [26].

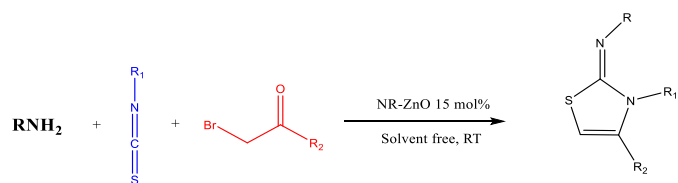


Fig. 3: Reaction of amines, alkylbromide, and isothiocyanates

III. COPPER OXIDE NANOPARTICLES (CUO NPs)

CuO NPs, recognized for their redox behavior and catalytic activity, have also been employed successfully [27-29]. A straightforward, very convenient method for the synthesis of 2-N-substituted benzothiazoles has been accomplished based on the use of nano copper oxide as a recyclable catalyst. The current tandem process provides straightforward access to a variety of 2-N-substituted benzothiazoles in good to excellent yield via the reaction of 2-iodo aniline with carbon disulfide and piperidine in the presence of KOH as a base and DMSO as a solvent under ligand-free conditions, Figure 4[30].

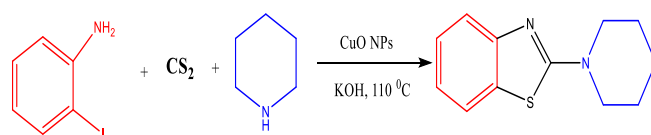


Fig. 4: Synthesis of benzothiazole derivatives using CuO NPs as a catalyst

In Figure 5, the utilization of copper nanoparticles/diorganyl dichalcogenides in activating direct carbon–hydrogen bond activation in thiazoles to afford 2-(organochalcogen)thiazoles. A detailed investigation of the catalytic system determined an integral role for the presence and the stoichiometric quantity of base to yield products. Substituents in the aromatic ring of the chalcogen bonded to the diorganyl dichalcogenides can be classified as electron-donating, electron-withdrawing, or neutral, and in all cases yield a product with one equiv. of base. However, when neutral substituents are on the aromatic ring of the chalcogen bonded to the diorganyl dichalcogenide, two equivalents of base were required to obtain the same yield. The justification for utilizing twice as much of the base in the reaction described in the attachment is due to the nature of the substituents on the aromatic ring of the chalcogen bonded to the diorganyl dichalcogenides. Catalytic systems of copper nanoparticles and diorganyl dichalcogenides enable direct carbon–hydrogen bond activation in thiazoles, which yields 2-organochalcogen-thiazole [31].

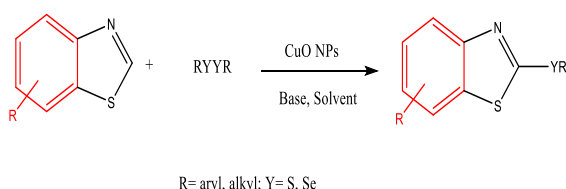


Fig. 5: Synthesis of benzothiazole derivatives using CuO NPs as a catalyst

IV. FERRIC OXIDE NANOPARTICLES (Fe₂O₃ NPs)

On the other hand, magnetically separable nanocatalysts are gaining importance in the eyes of researchers since they possess various advantages, including thermal stability, high surface area, reusability, biocompatibility, etc [32]. Among them, magnetically separable nano ferrites have been more widely employed in organic transformations because they can be cleanly isolated from the reaction mixture [33-40].

A new approach, in one-pot, a multicomponent reaction of tosylates, aryl aldehydes, and thiosemicarbazide, for the synthesis of hydrazinyl thiazole, using rust-iron-derived Fe₂O₃ NPs as a catalyst. It easily recovered the magnetic Fe₂O₃ NPs using an external magnet for the recycled catalytic studies of the catalyst four times under optimized reaction conditions, and the yield did not change very much in the four reaction phases combined, ranging between 92-95% (Figure 6) [41].

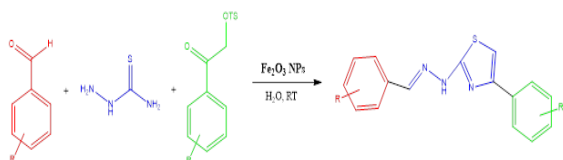


Fig. 6: Synthesis of hydrazinyl thiazoles using Fe₂O₃ NPs as a catalyst.

V. TITANIUM DIOXIDE NANOPARTICLES (TiO₂ NPs)

The surface acidity exhibited by TiO₂ NPs, particularly in their anatase form, makes them efficient photocatalysts. TiO₂ nanoparticles are commercially available materials that are non-toxic, economical, stable in moisture, and reusable, which has attracted significant scientific attention in recent years [42]. Typically, many similar applications of this nanoscale metal oxide as an active catalyst for green synthetic organic chemistry have been identified in the literature. However, the separation of these tiny titania particles from the reaction mixtures and the reusability of the catalysis process are difficult. This is a major limitation of the catalysis method for chemical reactions. Therefore, one of the big challenges of modern catalysis research involves the realization of titania-based materials with high catalytic

activity and ease of separation [43-48]. Safari et al. have been interested in developing a new, easy, and green method to synthesize 2-aminothiazoles via TiO₂ nanoparticles supported on the Fe₃O₄@ SiO₂ which can be used as an effective nanocomposite [49].

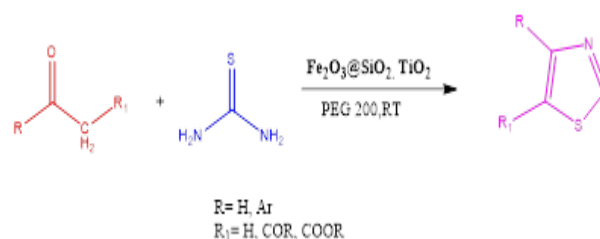


Fig.7: Synthesis of 2-aminothiazoles by Fe₃O₄@SiO₂.TiO₂ nanocomposite as a catalyst

A. Zirconium Dioxide Nanoparticles (ZrO₂ NPs)

Moreover, a principal drawback of nearly all of the currently implemented methods is that the catalysts are rendered inactive in the work-up step and cannot be accessed for re-use. In general, the search is ongoing for an ideal catalyst for the production of benzimidazoles, which includes operational simplicity, coupled with reusability and cost. Some recent reports on heterogeneous catalysts for the production of heterocyclic compounds have also been reported [50-54]. ZrO₂ NPs show both acidic and basic sites on their surfaces, and have shown high thermal stability, rendering them good amphoteric catalysts [55-56]. A novel and efficient catalyst based on nano-sized Co/Ce-ZrO₂ (25%) has emerged to synthesize 2- arylbenzothiazoles. This method has distinct advantages, including a simple and convenient procedure, easy purification of products, short reaction times, and a reusable catalyst [57].

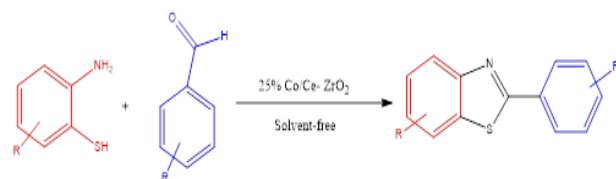


Fig. 8: Synthesis of 2-substituted-1H-benzimidazoles using 25 % Co/Ce-ZrO₂ as catalyst

VI. BISMUTH OXIDE NANOPARTICLES (Bi₂O₃ NPs)

Recently, bismuth (III) compounds have garnered significant interest in organic synthesis because of their high acidity, low toxicity, and relatively good stability. Sharma et al. describe a new strategy for synthesizing benzothiazole derivatives that involves refluxing at 60 °C, 2-

aminothiophenol with various aromatic aldehydes in ethanol with bismuth (III) oxide nanoparticles acting as a heterogeneous, non-toxic, and environmentally benign catalyst (Figure 9) [58].

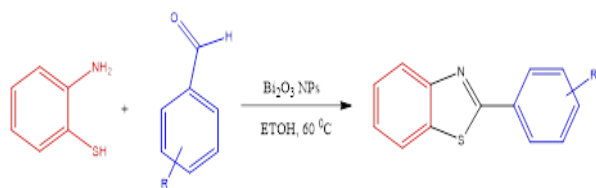


Fig 9: Synthesis of benzothiazole derivatives using Bi₂O₃ NPs as a catalyst

VII. CONCLUSION

Metal oxide nanoparticles (MONPs) have appeared as effective, recoverable, and sustainable catalysts for the synthesis of biologically important benzothiazole and thiazole derivatives. This review explains that MONPs, such as ZnO, TiO₂, Fe₂O₃, CuO, ZrO₂, and Bi₂O₃, can successfully address the inadequacies of classical approaches by delivering high catalytic selectivity, activity, and recyclability. Their utilization in one-pot, multicomponent reactions is consistent with green chemistry, leading to improved atom economy and by-product reduction. Examples of advantages for using MONPs as nanocatalysts include the outstanding efficiency of ZnO NPs, easy magnetic recovery of Fe₂O₃ systems, and cooperative effects in a variety of nanocomposite configurations. The implications of MONP catalysis signal a shift toward more efficient synthetic methods that are also environmentally friendly and further pave the way for their importance in the future of heterocyclic and medicinal chemistry. This field's forthcoming challenges and trends will likely incorporate a focus on environmentally sustainable practices for achieving greener synthetic methods, increasing the catalyst recyclability and separation simplicity of any subsequent post-synthetic procedures. Continued innovation in several forms and activity support of nanocatalysts will be necessary to enhance the effectiveness of reactions and reduce waste, especially toxic wastes, preserving both the scientific maturity and sustainability of the entire field for the sake of future researchers.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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