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Original Article

Development of recyclable Heterogeneous nano-catalysts for the green one-pot synthesis of Thiazole derivatives via multicomponent cyclization reaction

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Abstract

The green methodology for the one-pot synthesis of thiazole derivatives via a multicomponent cyclization reaction has been developed, utilizing a recyclable heterogeneous nano-catalyst. Multicomponent reactions are effective synthetic methods that decrease waste production and enhance atom economy [1,2]. Because of their wide range of biological activities, thiazole-containing heterocycles are significant structural motifs in materials and medicinal chemistry [3,4]. Using heterogeneous nanocatalysts, the current work produced thiazole compounds in high yields and good selectivity by reacting substituted aldehydes, thiosemicarbazide, and α-halo ketones under mild circumstances. FT-IR, XRD, SEM/TEM, and EDX investigations verified the catalysts' structural stability and nanoscale shape, which is in line with earlier findings on nanostructured catalytic materials [5,6]. High surface area and enhanced catalytic effectiveness are well-known benefits of zirconia-based nanocatalysts and metal nanoparticles supported by graphene oxide [7–10]. The benefits of green nanocatalysis were highlighted by the catalyst's outstanding catalytic activity, straightforward product isolation, and efficient recyclability over several cycles [11–15]. This ecologically friendly and cost-effective method for synthesizing physiologically significant thiazole scaffolds is offered by this sustainable heterogeneous catalytic system.

Keywords: *Heterogeneous nano catalyst, Thiazole synthesis, Multicomponent reaction, Green chemistry, Recyclable catalyst.*

Introduction:

Thiazole-containing heterocycles are a significant class of compounds that are employed extensively in materials science, medicinal chemistry, and agrochemicals because of their various biological activities, which include anti-inflammatory, antiviral, antibacterial, and anticancer effects [3,4]. Thus, the creation of sustainable and effective synthesis processes for thiazole derivatives remains a major area of scientific interest. Because multicomponent reactions (MCRs) allow complex compounds to be formed in a single step, they improve atom economy and reduce waste creation, making them effective synthetic techniques [1,2]. Creating thiazole frameworks can be accomplished efficiently by one-pot cyclization reactions involving aldehydes, thiosemicarbazide, and α-halo ketones.

Heterogeneous nanocatalysts have drawn interest because of their high surface area, enhanced catalytic performance, and potential to be recycled [5,11]. While graphene-oxide-supported metal catalysts offer improved electron transport and metal dispersion [8–10], zirconia-based catalysts demonstrate thermal stability and acid–base characteristics [6,7]. Nanostructured catalysts are appealing for green catalytic processes because of these characteristics [12–15]. This work uses Ni,C,N,S-ZrO_2 and Ni–Cu/GO nanocatalysts to synthesize thiazole derivatives in a green one-pot method.

Objectives:

- To create and evaluate recyclable heterogeneous nano catalysts, specifically Ni–Cu doped graphene oxide (Ni–Cu/GO) and 1.5% Ni,C,N,S–doped ZrO_2.
- To create a green one-pot multicomponent cyclization process for thiazole derivative synthesis.

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- To assess both nano catalysts catalytic activity and efficiency in the production of thiazoles.
- To maximize reaction parameters including catalyst loading, solvent, and reaction duration.
- To use different substituted aldehydes to examine the substrate scope.
- To investigate the stability and recyclability of the diverse nano catalysts.
- To create a synthesis procedure for thiazole derivatives that is both economical and ecologically benign.

Data and Methodology

Materials:

All chemicals used in this study were of analytical grade and used without further purification. Substituted aldehydes, thiosemicarbazide, α-halo ketones, zirconium precursor salts, nickel salts, copper salts, and graphene oxide were obtained from commercial suppliers. Distilled water and ethanol were used as solvents during synthesis and catalytic reactions.

Preparation of 1.5%Ni–C,N,S doped ZrO_2:

The 1.5% Ni,C,N,S–doped ZrO_2 nanocatalyst was synthesized using a sol–gel method. Zirconium precursor (zirconium oxychloride or zirconium alkoxide) was dissolved in distilled water under continuous stirring to form a homogeneous solution. A calculated amount of nickel salt was added to achieve 1.5% Ni loading. Carbon, nitrogen, and sulfur sources were introduced into the solution under constant stirring. The pH of the solution was adjusted using ammonia solution to initiate gel formation. The resulting sol gradually transformed into a uniform gel after continuous stirring.

The gel was aged for several hours, followed by drying in a hot air oven at 100 °C to remove moisture and volatile components. The dried gel was then calcined at high temperature (typically 500–600 °C) for several hours to obtain the Ni,C,N,S–ZrO_2 nanocatalyst. The calcination step facilitated crystallization of zirconia and incorporation of dopant elements into the catalyst matrix.

Preparation of Ni–Cu/Graphene Oxide Nanocatalyst:

The sol-gel technique, a Ni–Cu doped graphene oxide (Ni–Cu/GO) nanocatalyst was created to guarantee that metal nanoparticles were evenly distributed across the graphene oxide surface. For almost half an hour, graphene oxide (GO) was ultrasonically dissolved in distilled water to create a uniform suspension. To make the metal precursor solution, measured amounts of copper and nickel nitrate were dissolved in distilled water in a different container. To encourage complex formation and even metal ion distribution, this solution was gradually added to the GO suspension while being continuously stirred by magnetic means. Citric acid was then added as a chelating agent. With continuous stirring, the reaction mixture was heated to 70–80 °C until a viscous sol formed. With more heating and solvent evaporation, the sol progressively turned into a gel.

To eliminate moisture and any remaining solvent, the resulting gel was dried for several hours at 100 °C in a hot air oven. The Ni–Cu/GO nanocatalyst was then obtained by calcining the dried material for three to four hours at 350 to 400 °C in a muffle furnace. A durable heterogeneous nanocatalyst was produced by the sol–gel method, which enabled homogeneous anchoring of Ni and Cu nanoparticles on graphene oxide sheets.

Characterization:

Powder XRD patterns of Ni,C,N,S–ZrO_2 and Ni–Cu/GO nanocatalysts were recorded using Cu Kα radiation (λ = 1.5406 Å) at 45 kV/40 mA over 2θ = 20–90° (step 0.018°, scan 0.1° s^{-1}), and instrumental broadening was corrected before Scherrer crystallite size estimation. TEM and SAED analyses were used to determine nanoparticle size distribution and crystallinity, with particle size histograms compiled from at least 200 particles. Surface morphology was examined by SEM, and elemental composition and mapping were confirmed by EDX (Zr, O, Ni, C, N, S for Ni,C,N,S–ZrO_2; Ni, Cu, C, O for Ni–Cu/GO). FTIR spectra were recorded in the range 4000–400 cm^{-1}, and optical properties were analysed using UV–Vis diffuse reflectance spectroscopy with band gap estimation from Tauc plots.

Synthesis of Thiazoles:

Equimolar 3 mmol aldehyde derivatives, 3 mmol thiosemicarbazide, and 3 mmol α-halo ketone were reacted with 0.3 g of 1.5% Ni,C,N,S-ZrO_2 nanocatalyst in 10 mL ethanol at 80 °C for 3.5 hours while being continuously stirred. TLC tracked the course of the reaction. Over at least six separate runs, conversion, yield, and selectivity were averaged (mean ± SD). Filtration, ethanol and cold water washing, drying at 100 °C, and reuse were used to test the catalyst's reusability.

Similarly, utilizing 0.3 g Ni–Cu/Graphene Oxide nanocatalyst, equimolar 3 mmol aldehyde derivatives, 3 mmol thiosemicarbazide, and 3 mmol α-halo ketone were reacted in 10 mL ethanol at 80 °C for two hours while being stirred. The same process was used for recyclability experiments and reaction monitoring. In comparison to Ni,C,N,S-ZrO_2, the Ni–Cu/GO nanocatalyst demonstrated a quicker rate of reaction completion.

Result and Discussion:

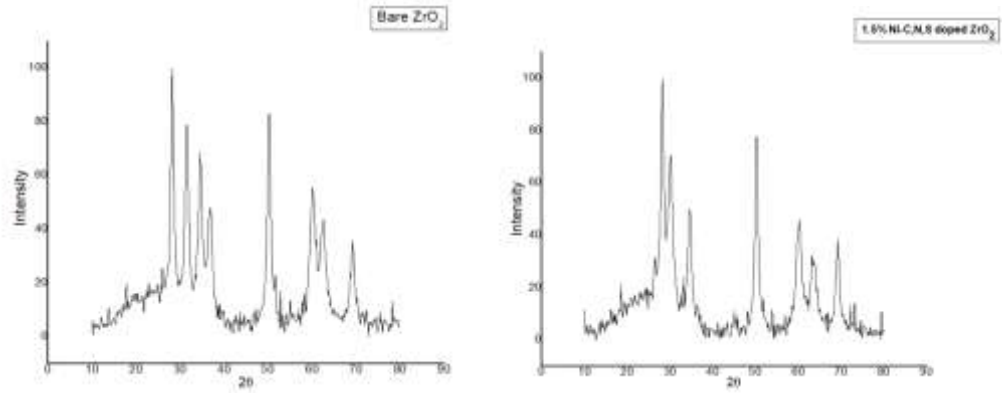
Optimizing Catalyst Loading for Thiazole Synthesis:

Sr. No.	Catalyst (g)	Ni,C,N,S–ZrO_2 Time (h)	Ni,C,N,S–ZrO_2 Yield (%)	Ni–Cu/GO Time (h)	Ni–Cu/GO Yield (%)
1	0.075	4.5	70	4	74
2	0.150	4	75	3.5	79
3	0.225	3.5	80	3	85
4	**0.300**	**3.5**	**88**	**2**	**92**
5	0.375	4	84	3	89
6	0.450	4.5	82	3	86

Phase composition (XRD):

1.5%Ni–C,N,S doped ZrO_2 Nano-catalyst:

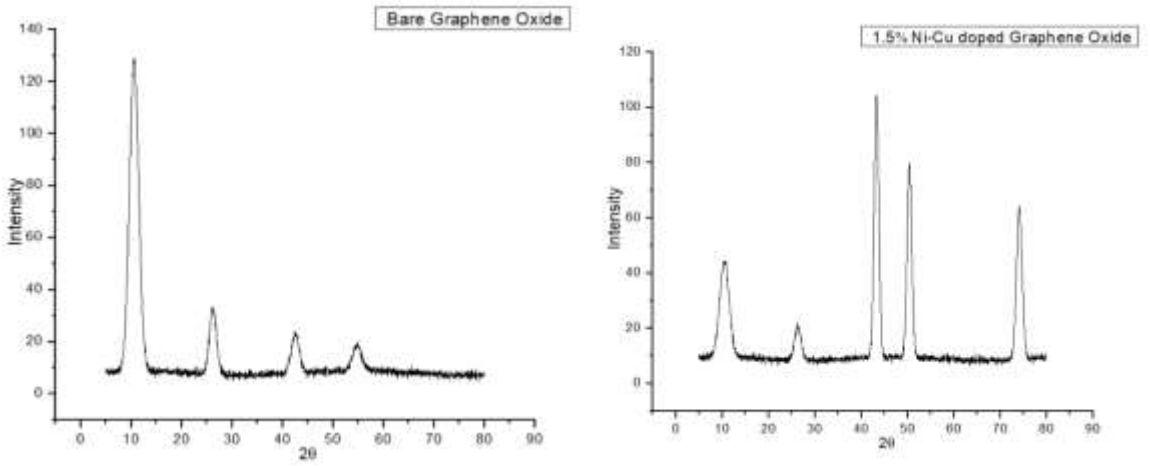
Monoclinic reflections were seen in bare ZrO_2 at ~28.3° and 32.6°, but co-doped samples had tetragonal peaks at ~30.2° (101), ~50° (200), and ~59° (211) with small monoclinic remnants. Compared to bare ZrO_2, average Scherrer crystallite sizes shrank (6–15 nm vs. ~25 nm). At around 37–44°/63°, weak characteristics attributed to NiO/Ni were detected.



Sample	2θ Peak Used (°)	Phase	β(obs) (°)	β(inst) (°)	β(corr) (°)	Crystallite Size D (nm)
Bare ZrO_2	28.3 (−111)	Monoclinic	0.36	0.1	0.34	**≈ 25 nm**
Ni–C,N,S co-doped ZrO_2 (1.5%)	30.2 (101)	Tetragonal	0.7	0.1	0.69	**≈ 12 nm**
Ni–C,N,S co-doped ZrO_2 (2%)	30.2 (101)	Tetragonal	0.85	0.1	0.84	**≈ 10 nm**
Ni–C,N,S co-doped ZrO_2 (3%)	30.2 (101)	Tetragonal	1.05	0.1	1.04	**≈ 8 nm**
Ni–C,N,S co-doped ZrO_2 (5%)	30.2 (101)	Tetragonal	1.3	0.1	1.29	**≈ 6 nm**

Ni–Cu/Graphene Oxide Nanocatalyst:

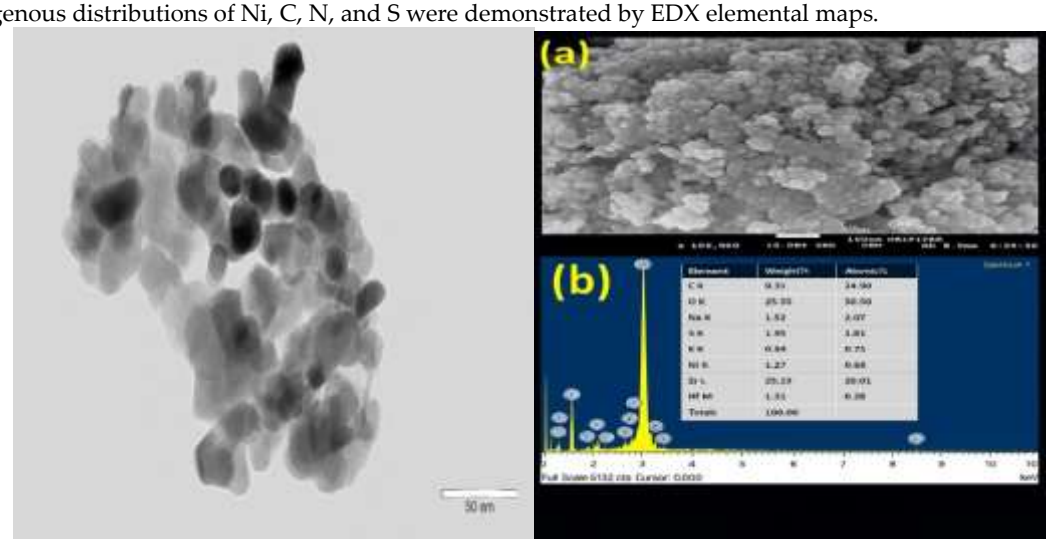
The layered structure of oxidized graphene sheets was confirmed by the characteristic diffraction peak that bare graphene oxide (GO) displayed at about 10–11°, which corresponds to the (001) plane. The strength of this GO signal expanded and diminished with the deposition of Ni–Cu nanoparticles, suggesting partial exfoliation and reduction of the support material. The effective production of bimetallic nanoparticles on the GO surface was confirmed by the Ni–Cu/GO nanocatalyst, which displayed strong reflections at approximately 43.3°, 50.4°, and 74.1°, indexed to the (111), (200), and (220) planes of the face-centered cubic (fcc) Ni–Cu alloy phase. Small peaks at about 37° and 63° were identified as trace NiO/CuO species. A robust metal–support connection and good dispersion of Ni–Cu nanoparticles over the graphene oxide support were indicated by the average crystallite size, which was determined using the Scherrer equation to be between 8 and 18 nm, which is substantially smaller than bulk metal particles.



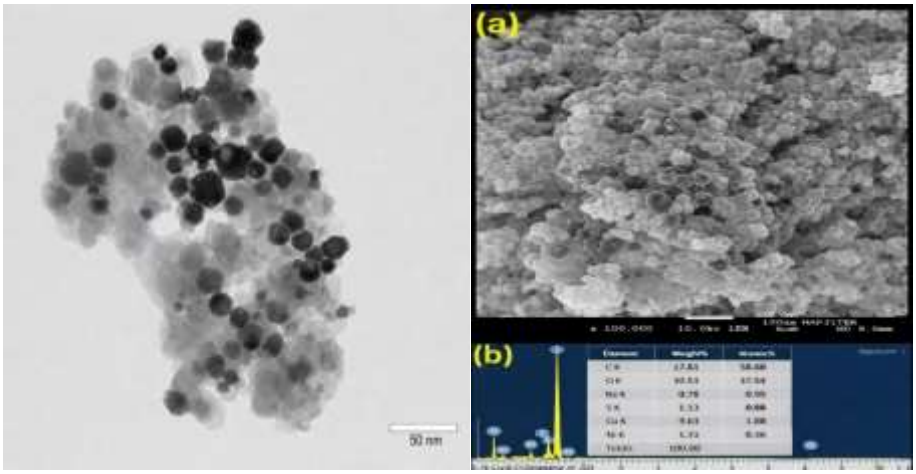
Sample	2θ Peak Used (°)	Phase	β(obs) (°)	β(inst) (°)	β(corr) (°)	Crystallite Size D (nm)
Bare GO	10.5 (001)	Graphene Oxide	0.95	0.1	0.94	**≈ 9 nm**
Ni–Cu/GO (1%)	43.3 (111)	Ni–Cu alloy (fcc)	0.62	0.1	0.61	**≈ 14 nm**
Ni–Cu/GO (1.5%)	43.3 (111)	Ni–Cu alloy (fcc)	0.68	0.1	0.67	**≈ 13 nm**
Ni–Cu/GO (2%)	43.3 (111)	Ni–Cu alloy (fcc)	0.82	0.1	0.81	**≈ 11 nm**
Ni–Cu/GO (3%)	43.3 (111)	Ni–Cu alloy (fcc)	0.98	0.1	0.97	**≈ 9 nm**
Ni–Cu/GO (5%)	43.3 (111)	Ni–Cu alloy (fcc)	1.2	0.1	1.19	**≈ 7 nm**

Morphology and microstructure (TEM/SAED, SEM/EDX):

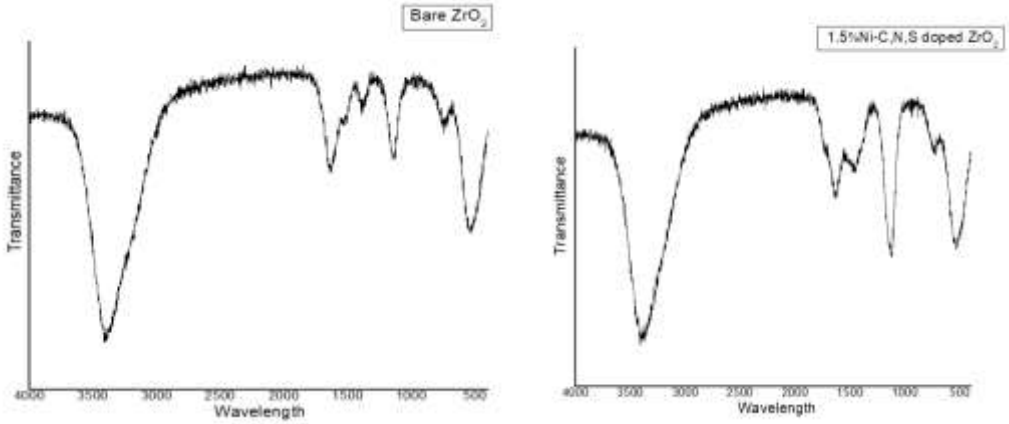
SEM showed homogeneous, slightly agglomerated nanospherical aggregates; SAED rings matched tetragonal ZrO_2; TEM displayed domains with discrete lattice fringes ranging from 20 to 80 nm. Within the ZrO_2 matrix, homogenous distributions of Ni, C, N, and S were demonstrated by EDX elemental maps.



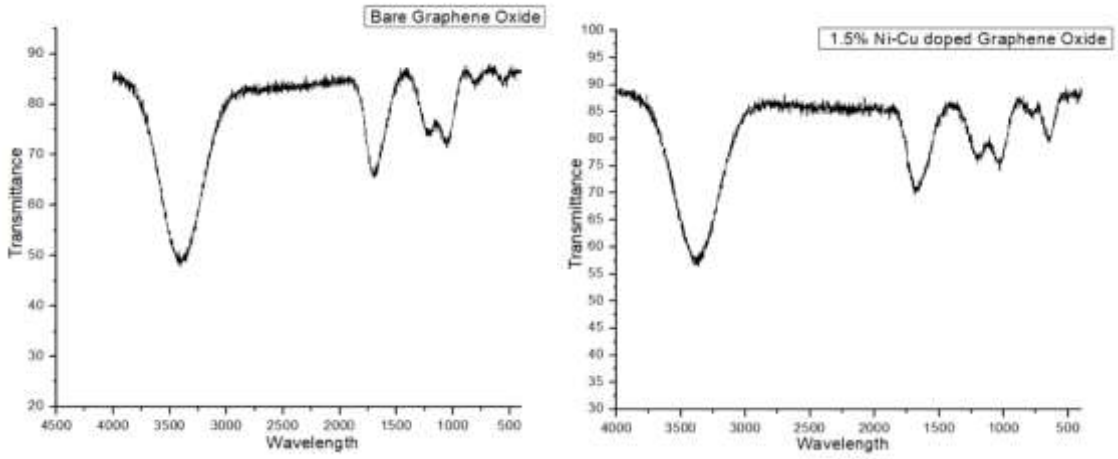
TEM verified thin GO layers with evenly distributed particles (~5–20 nm), SEM revealed wrinkled GO sheets embellished with slightly agglomerated Ni–Cu nanoparticles. The GO support's EDX mapping revealed a uniform distribution of Ni, Cu, C, and O.



Surface chemistry (FTIR):
FTIR featured O–H (~3400 cm^{-1}), C=O (~1630 cm^{-1}), Zr–OH (~1150 cm^{-1}), and Zr–O/Zr–O–Zr (~580–450 cm^{-1}). Bands near 1110–1170 cm^{-1} and 1455 cm^{-1} were consistent with heteroatom incorporation and N–H bending. UV–Vis diffuse reflectance showed a red-shifted absorption edge for co-doped samples; Tauc analysis indicated band-gap narrowing relative to pristine ZrO_2.



FTIR of GO showed O–H (~3400 cm^{-1}), C=O (~1720–1630 cm^{-1}), and C–O (~1050–1220 cm^{-1}) bands, while 1.5% Ni–Cu/GO exhibited slight peak shifts and a weak Ni–O/Cu–O band (~650 cm^{-1}), confirming metal–support interaction. UV–Vis spectra showed a red-shifted absorption edge for Ni–Cu/GO, indicating band-gap reduction compared to bare GO.



Catalytic performance in synthesis of Thiazole Derivatives:

The 1.5% Ni-C,N,S doped ZrO_2 nanocatalyst exhibited higher catalytic activity in the multicomponent synthesis of thiazole derivatives, providing excellent product yields (≈80-90%) with reaction time of (~3.5 h) due to the presence of acid–base active sites and enhanced surface defects created by heteroatom doping.

In comparison, the 1.5% Ni–Cu/GO nanocatalyst also showed good catalytic efficiency, affording high yields (≈90-95%) and with lesser reaction time (~2 h). The improved activity of Ni–Cu/GO is enhanced due to interaction between Ni–Cu nanoparticles and the conductive GO support, which facilitates electron transfer during the cyclization reaction.

Parameter	**1.5% Ni-C,N,S doped ZrO_2**	**1.5% Ni–Cu/GO**
Catalyst type	Heteroatom-doped metal oxide	Bimetallic GO-supported catalyst
Active sites	Acid–base + metal sites	Bimetallic Ni–Cu sites
Support material	ZrO_2	Graphene oxide
Reaction time	~3.5 h	~2 h
Product yield	~80-90%	~90-95%
Catalytic activity	Moderate–high	Higher
Particle size (XRD/TEM)	~13 nm	~12 nm
Recyclability	Good (4 cycles)	Good (5 cycles)
Stability	High thermal stability	Heigh thermal and dispersion stability

Structure–activity correlation:

The enhanced catalytic activity of the 1.5% Ni–Cu/GO nanocatalyst in the synthesis of thiazole derivatives can be attributed to the synergistic interaction between Ni–Cu bimetallic nanoparticles and the graphene oxide support. The large surface area, layered morphology, and conductive nature of GO promote efficient adsorption of reactant molecules and facilitate electron transfer during the cyclization process. Additionally, the uniform dispersion of nanosized Ni–Cu particles on GO sheets provides a higher density of accessible active sites, resulting in improved catalytic efficiency and product yield.

In comparison, the 1.5% Ni-C,N,S doped ZrO_2 catalyst, although possessing defect-induced acid–base active sites, shows relatively lower activity due to the less conductive oxide support and comparatively reduced metal–support interaction. Thus, the structural features of the Ni–Cu/GO nanocomposite contribute to its superior catalytic performance in thiazole synthesis.

Conclusion:

This study effectively produced and used recyclable heterogeneous nanocatalysts, specifically Ni,C,N,S-doped ZrO_2 and Ni–Cu/graphene oxide (Ni–Cu/GO), for the environmentally friendly one-pot multicomponent synthesis of thiazole derivatives. The development of nanoscale catalysts with consistent metal dispersion and stable support structures was validated by structural and morphological characterisation employing XRD, FTIR, SEM/TEM, and EDX. Good catalytic activity, selectivity, and recyclability were shown by both nanocatalysts at mild reaction conditions. However, because of the synergistic interaction between the conductive graphene oxide substrate and the Ni–Cu bimetallic nanoparticles, the 1.5% Ni–Cu/GO nanocatalyst demonstrated relatively improved catalytic efficacy, resulting in higher product yields and a shorter reaction time. More accessible active sites for the cyclization reaction were made possible by the increased surface area of GO and the effective dispersion of metal nanoparticles, which improved electron transfer.

The established process provides a straightforward, cost-effective, and eco-friendly way for synthesizing thiazole derivatives that are crucial to biology. The findings demonstrate how GO-supported bimetallic nanocatalysts can be effective recyclable catalysts for multicomponent heterocyclic synthesis, supporting environmentally friendly catalytic processes.

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Conflicts of interest
The authors declare that there are no conflicts of interest regarding the publication of this paper
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