

CoFe₂O₄ NANOCATALYST MEDIATED MICROWAVE ASSISTED ONE-POT SYNTHESIS OF 2-(2-(ARYL METHYLIDENE) HYDRAZINYL 1,3-THIAZOL-4-(5H)-ONE DERIVATIVES

Komal V. Takankhar and Ayesha Durrani✉

Department of Chemistry, Dr. Rafiq Zakaria College for Women,
Chhatrapati Sambhajinagar-431001, Maharashtra, India.

✉Corresponding author: drayeshanuzhat101@gmail.com

ABSTRACT

A simple one-pot, two-step synthesis and facile protocol have been successfully developed for a series of 2-(2-(aryl methylidene) hydrazinyl)-1,3-thiazol-4(5H)-one derivatives. Substituted aromatic aldehydes, thiosemicarbazide and α -haloesters were reacted in the presence of a CoFe₂O₄ nanocatalyst to afford novel thiazolidine derivatives. This strategy operates under mild conditions, producing highly pure products (90–97%) without the need for chromatographic separation. The sol-gel synthesized CoFe₂O₄ nanocatalyst accelerates reaction rates, reduces completion time and enhances yields, while maintaining excellent reusability. Beyond its synthetic efficiency, this work carries broader significance for the field of coordination and medicinal chemistry: It demonstrates how nanocatalysts can be harnessed to design greener, more sustainable synthetic routes, thereby reducing reliance on hazardous solvents and energy-intensive processes. Such advances not only contribute to academic progress in catalysis and heterocyclic chemistry but also provide practical pathways for scalable, eco-friendly drug discovery and material development. By aligning with the principles of green chemistry, this study directly supports Sustainable Development Goal-12: Responsible Consumption and Production, underscoring its potential impact on both scientific innovation and environmental stewardship.

Keywords: One-Pot Synthesis, Microwave-Assisted, Hydrazinyl 1,3-thiazol -4(5H)-one, CoFe₂O₄ Nanocatalyst, Catalyst Reusability, EtOH Solvent etc.

RASAYAN J. Chem., Vol. 19, No.4, 2026

INTRODUCTION

Nanostructured spinel ferrites, particularly cobalt ferrite (CoFe₂O₄), have attracted sustained attention due to their versatile applications in magnetic fluids, high-density data storage, and biomedical fields, owing to their mechanical hardness, high coercivity and moderate saturation magnetisation.¹ The magnetic and dielectric properties of CoFe₂O₄ nanoparticles are strongly influenced by synthesis methods, chemical composition, and post-synthesis heat treatment, making them a promising candidate for tailored functional materials.² Their inherent chemical stability, large magnetic anisotropy and ease of recovery through magnetic separation further enhance their utility in catalysis and environmental remediation. Despite these advantages, a critical research gap exists in developing eco-friendly catalytic systems that simultaneously improve reaction efficiency and reduce environmental impact. Traditional synthetic methods for heterocyclic compounds, such as thiazolidinones and thiazoles, often rely on energy-intensive processes or hazardous reagents. Addressing this challenge, our study explores the use of CoFe₂O₄ nanocatalysts in the synthesis of thiazolidinone derivatives, aiming to establish a sustainable, high-yield, and reusable catalytic protocol.^{3,4,5} This approach not only advances synthetic efficiency⁶ but also aligns with the principles of green chemistry, thereby contributing to Sustainable Development Goal (SDG) 12: Responsible Consumption and Production by promoting cleaner production pathways and reducing chemical waste. Thiazolidinones and thiazoles are of considerable pharmaceutical interest due to their broad biological activities, including antimicrobial, anticancer, anti-inflammatory and antifungal properties. Recent studies highlight innovative synthesis strategies such as microwave-assisted reactions and bio-based nanocatalysts, which significantly enhance yields (up to 97%) and reduce reaction times compared to conventional methods. Integrating CoFe₂O₄ nanocatalysts into these processes offers a dual

advantage: improved catalytic performance and sustainable recovery, thereby optimising thiazolidinone production for potential therapeutic applications.⁷ Furthermore, thiazole derivatives continue to demonstrate diverse pharmacological relevance⁸, serving as antimicrobial agents, anticancer drugs (tiazofurin) and industrial additives such as vulcanising accelerators.⁹ Their expanding role in novel therapeutic applications-including antithrombotic agents, DNA gyrase inhibitors and treatments for conditions such as HIV, hypertension and schizophrenia underscores the importance of developing efficient and sustainable synthetic routes.¹⁰ By bridging the gap between nanocatalysis and heterocyclic drug development, this study contributes both to academic discourse in catalysis and to practical advances in pharmaceutical chemistry.¹¹

EXPERIMENTAL

Materials and Methods

Spectral data such as ¹H NMR and ¹³C NMR Obtained in Bruker Avance spectrometer Neo 500 (DRX)MHz, NMR spectrometer, XRD recorded in Anton parr Dynamic 500, Buchi melting point apparatus B-540 B.V.CHI used for Melting points determination, All the chemicals Cobalt nitrate (0.1 mol), ferric nitrate, (0.2 mol) citric acid (0.3 mol) ammonium solution (adjust up to pH 6-7), Thiosemicarbazide (0.01 mol), sub. benzaldehyde (0.01 mol), EtOH (15 mL). Required reagents: chemicals acquired from Sigma-Aldrich and Merck Private Limited. Synthesized CoFe₂O₄ NPs (10 mol %) were taken after completing the reaction to check the reaction condition; aluminium foil plates covered with silica gel using hexane: ethyl acetate (7:3) as the solvent system ratio used for thin layer chromatography (TLC), catalyst separated by using an external permanent magnet.

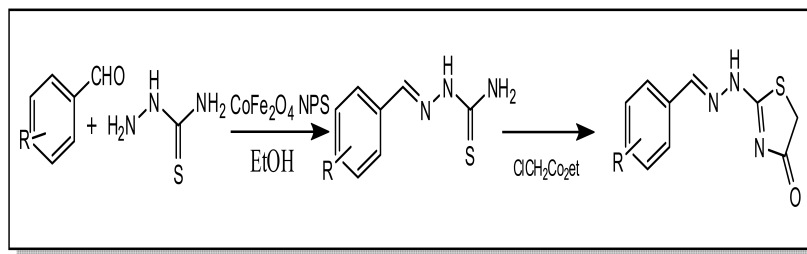
General Procedure of CoFe₂O₄ Nanocatalyst

For the preparation of CoFe₂O₄ NPs, Fe(NO₃)₃ · 9 H₂O (0.2 mol) & Co (NO₃)₂ · 6 H₂O (0.1 mol), citric acid (0.3 mol), and ammonia solution was taken in stoichiometric amounts of ferric nitrate and cobalt nitrate dissolved in dist. H₂O, the resulting solution was stirred constantly at RT until a uniform solution formed. Citric acid added to this solution in 1:2:3 molar ratio, the solution heated up to 80-90 °C under constant stirring, ammonia solution added up to PH 7, this mixture heated at 80-90°C until viscous gel was formed, in hot air oven prepared gel dried at 100-120°C for 1 hrs. to remove excess moiety and obtained dried compound was grinded in mortar and pestle up to fine powder obtained after that calcination process done in muffle furnace upto 700°C for 3 hrs. to obtained black crystalline powder of CoFe₂O₄ nanoparticles, synthesized nanocatalyst has been characterized and used for catalyst in organic synthesis reaction.

General Procedure of 2-(2-(Aryl methylidene) hydrazinyl)1,3- thiazol -4(5H)-one

Thiosemicarbazide (0.01mol), sub. benzaldehyde (0.01mol) in EtOH (15ml) furthermore to this synthesized CoFe₂O₄ nanocatalyst(10 mol %) has been added the prepared solution heated at 80°C up to 1 h., to ensure the complete consumption of benzaldehyde, next to the added of ethyl chloroacetate (0.01 mol) in to above mixture, progress of reaction checked by TLC, reaction quenched by addition of ice cold water leading to solid formation, using External permanent magnet catalyst removed from product, after filtration obtained purification of via recrystallization using ethanol desired 90% yield of 2-(2-(Aryl methylidene) hydrazinyl) 1,3-thiazol -4(5H)-one was obtained in pure form for spectral analysis.

Experimental Reaction



Scheme-I: One Pot Preparation of 2-(2-(aryl methylidene) hydrazinyl)1,3- thiazol -4(5H)-one using CoFe₂O₄ Nanocatalyst

RESULTS AND DISCUSSION

The derivatives of 2-(2-(aryl methylidene)hydrazinyl)-1,3-thiazol-4(5H)-one were synthesized in high yield through an optimised one-pot protocol employing CoFe_2O_4 nanoparticles as a catalyst. Structural confirmation of the products was achieved using ^1H -NMR and ^{13}C NMR spectroscopy, while the nanocatalyst was rigorously examined by XRD, SEM and TEM, verifying its crystalline framework and nanoscale features. The study highlights two key contributions: first, the development of a straightforward and efficient synthetic route for thiazolidinone derivatives; second, the demonstration of CoFe_2O_4 nanoparticles as a recyclable and sustainable catalyst that accelerates reactions without being consumed. By integrating nanotechnology with heterocyclic synthesis, this work advances the field of green chemistry and provides valuable insights for designing eco-friendly catalytic systems. The findings not only enrich the existing body of knowledge on nanocatalyst applications but also open pathways for future exploration in medicinal chemistry and materials science, reinforcing the role of nanotechnology in sustainable innovation.

Characterization of Synthesized CoFe_2O_4 Nanocatalyst

XRD (X-ray Diffraction)

Synthesized CoFe_2O_4 nanoparticles exhibits excellence diffraction peaks at 2θ values coordinating with (111), (220), (311), (400), (422), (511), (440), (533), and (622) crystallographic planes, among these reflections, the (311) plane exhibits the highest intensity, shown in (Fig-1) that this plane is preferred orientation of the CoFe_2O_4 nanoparticle for the single phase cubic spinel-type composition of CoFe_2O_4 NPs was well matched with JCPDS Card No. 22-1086, then confirming successful formation, the sharp and well-defined diffraction peaks shown fine crystallinity of synthesized nanoparticles. Overall, the XRD analysis confirms that the prepared nanoparticle possesses a well-defined cubic spinel structure with good crystallinity and the nanoscale particle size confirms the successful formation of CoFe_2O_4 nanoparticles.

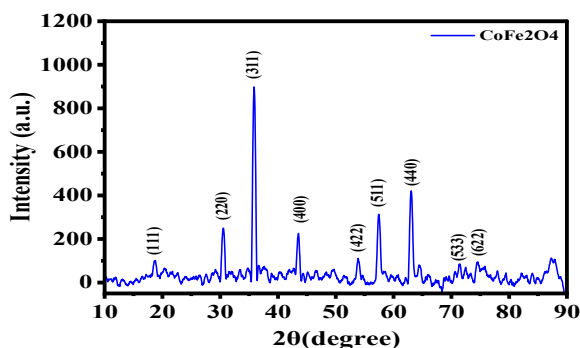


Fig.-1: XRD Spectrum of Synthesized CoFe_2O_4 Nanoparticle

SEM (Scanning Electron Microscopy)

Synthesized CoFe_2O_4 nanoparticles surface morphological Characteristics estimated using Scanning Electron Microscopy, Sem micrographs indicate the formation of nanoparticles with irregular but mostly quasi-spherical morphology. Particle size ranges from 39 nm to 56nm; most of the nanoparticles appeared in 40-55nm range, showing that the synthesized particles were in the nanometer scale. The obtained characteristics suggest that nanoparticles possess rough surface texture and porous aggregated structures; this provides higher surface area. Such structural arrangements are beneficial for catalytic applications. The SEM micrographs (Fig.-2) confirm the formation of agglomerates with usual size of particle size between 39 nm and 56 nm, showing good nanoscale morphology suitable for catalytic applications.

TEM (Transmission Electron Microscopy)

The TEM analysis shown that successful synthesis of CoFe_2O_4 nanoparticle with nanoscale dimensions 20-80 nm, irregular morphology and significant agglomeration images provides a high surface to volume ratio, which is beneficial to catalytic applications, then this Selected area electron diffraction (SAED) image of the synthesized CoFe_2O_4 NPs exhibits a series of bright, discrete diffraction spots arranged in a

symmetric pattern, indicating the presence of a highly crystalline structure, the sharpness and intensity of these spots suggests that the nanoparticles possess good crystallinity with well-defined crystalline spinel structure with high crystallinity as shown in (Fig.-3).

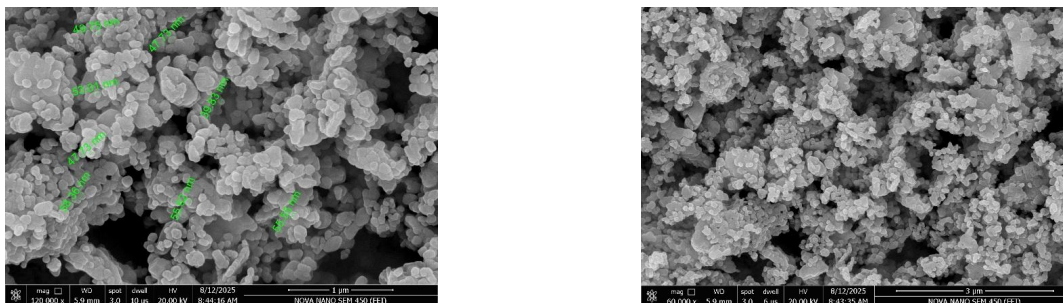


Fig.-2: SEM Images of Synthesized CoFe_2O_4 Nanoparticle

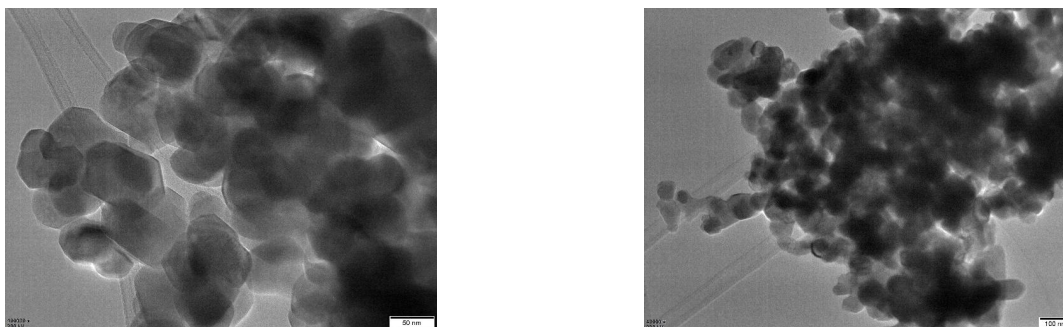


Fig.-3: TEM Images of Synthesized CoFe_2O_4 Nanoparticle

Spectral Data of Synthesized Thiazole derivatives

2-(2-(4-chloromethylidene) hydrazinyl),1,3- thiazol -4 (5H)-one [W2]

FT-IR(ν_{max} , cm^{-1}): 3320(N-H stretching), 3060 (Aromatic CH stretching), 2925(CH Stretching), 1716 (C=O), 1600(C=N) (C=C), 1510(Ar), 1220(C-N), 720(C-S), 745(C-Cl)

^1H NMR (DMSO- d_6 δ , ppm): 11.9(s, 1H, NH), 7.2-8.2(m, Aromatic-H), 4.5(s, d-1H, CH), 3.2-3.4(m, 2H, CH_2)

^{13}C NMR spectrum, (DMSO- d_6 δC , ppm): 173.06(C=O), 165.66, 154.96(C-N, C=N), 139.33, 137.98, 131.18, 129.88, 128.85(Ar-C), 60(CH), 33(CH_2 , C-2).

Mass spectrum: m/z = 257

2-(2-(4-methoxymethylidene) hydrazinyl) 1,3-thiazol -4 (5H)-one[X2]

IR spectrum(ν_{max} , cm^{-1}): 3278(N-H), 3075(Ar-C-H), 2890(C-H), 1700(C=O), 1620(C=N), 1560(Aromatic C=C), 1250 (C-O), 1090 (C-N), 760(C-S)

^1H NMR (DMSO- d_6 δ , ppm): 11.89(s, 1H, N-H), 7.2-8.0(d, 1H, Aromatic-H), 3.95-4(s, 1H, CH(C-2))

^{13}C NMR spectrum, (DMSO- d_6 δC , ppm): 161-158(C=O), 128.4-114.9(C-C, S-N), 33(CH_2 , C-2).

Mass spectrum: m/z = 217

2-(2-(4-hydroxymethylidene) hydrazinyl) 1,3-thiazol -4 (5H)-one[S2]

IR spectrum (ν_{max} , cm^{-1}): 3300(ph-OH stretching), 3165(N-H stretching), 3050(Ar C-H), 1715(C=O), 1630(C=N), 1567(C=N), 1280(C-N), 790(C-S)

^1H NMR (DMSO- d_6 δ , ppm): 11.65(s, 1H, NH), 9.95 (s, 1H, CH_2), 7.5-8.2 (s, 2H, CH_2 , Ar-H), 6.56(ph-OH) 6.4(s, 2H), 3.86(s, 2H)

^{13}C NMR spectrum, (DMSO- d_6 δC , ppm): 194.71(C=O), 159.79-156(C=N), 135.43, 132.13, 129.37, 126.13, 115.58(C-C, C=C)

Mass Spectrum: m/z = 235

2-(2-(4-dimethylmethylidene) hydrazinyl),1,3- thiazol -4 (5H)-one[R2]

IR spectrum: (ν_{max} , cm^{-1}): 3255(N-H stretching), 3080(Aromatic-CH), 2900(CH), 1700(C=O), 1620(C=N), 1560(Ar C=C), 1450, 1320 (C-N), 1130(C-S), 845(Ar C-H).

^1H NMR (DMSO- d_6 δ , ppm): 8.14(s,1H, NH), 7.59-7.51(2H, dd), 4.16(s,2H, C-2), 3.6(q,2H, C-5), 2.9-2.5(2H, dd, C-5), 1.07(3H, d)

^{13}C NMR spectrum, (DMSO- d_6 δC , ppm):152.08(C=O),151(C=N),128-122(Ar-C, C=C), 39 (CH₂, C-2)

Mass spectrum: m/z = 262.33

2-(2-(3,4-dimethoxymethylidene) hydrazinyl)1,3-thiazol-4(5H)-one[T2]

IR(ν_{max} , cm^{-1}):

3250(NH),3045(AromaticCH),2930(OCH₃),1700(C=O),1615(C=N),1550(AromaticC=C),1255(Ar-O-CH₃),1130(C-O),750(C-S)

^1H NMR (DMSO- d_6 δ , ppm):11.8(s,1H),7.8-7.0(m,5H),3.9(s,2H, C-2),3.8-3.5(s,2H, C-5)

^{13}C NMR spectrum, (DMSO- d_6 δC , ppm):175(C=O),135-140,128-130,55-60(CH₂, C-2) (38 CH₂, C-5).

Mass spectrum: m/z =279

Table-1: - Synthesis of 2-(2-(Aryl methylidene) hydrazinyl) 1,3-thiazol -4 (5H)-one using CoFe₂O₄ Nanocatalyst

S. No.	Entry	Conventional mode, Time(h)	Yield%	Microwave mode, Time(min)	Yield%
1	W2	5	79	25	85
2	X2	5	82	20	90
3	S2	4	75	25	88
4	R2	5	69	20	92
5	T2	4	70	30	87

Table-2: Selection of the Catalyst

S. No.	Catalyst	Reaction time(h/min)	Yield %
1	Net reaction	24 h	Trace
2	cyclodextrin	8h	52
3	Glycine	5h	56
4	CoFe ₂ O ₄	35	90

Catalyst Reusability

CoFe₂O₄ nanocatalysts reveal very good reusability because of their magnetic separability and structural robustness. It can be effectively reused and recovered for several cycles with only a minor reduction in catalytic activity, making it a cost-effective catalyst and environmentally friendly for organic synthesis, especially in heterocyclic compound formation. The results show that the CoFe₂O₄ nanocatalyst retained remarkable catalytic activity even after many cycles. The facile magnetic recovery, combined with sustained catalytic performance over various cycles, highlights the potential of the CoFe₂O₄ nanocatalyst as an efficiently reusable for heterogeneous catalyst for green and sustainable chemical processes.

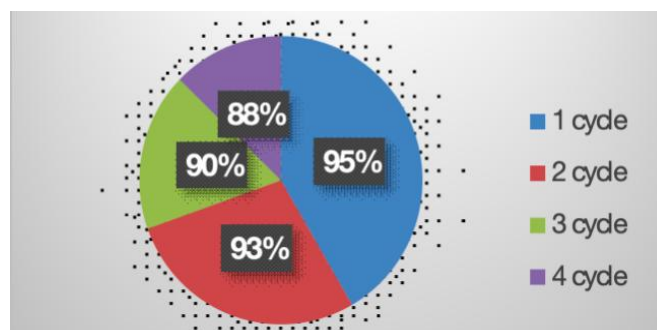


Fig.-4: CoFe₂O₄ Nanocatalyst Reusability Cycle

CONCLUSION

The CoFe₂O₄ nanocatalyst was prepared using the sol-gel method and thoroughly characterised by XRD, SEM and TEM, confirming its nanoscale structure and stability. Its application in the one-pot synthesis of 2-(2-(aryl methylidene)hydrazinyl)-1,3-thiazol-4(5H)-one derivatives yielded products of high purity, as

validated by ^1H -NMR, ^{13}C NMR, and mass spectroscopy. The catalyst accelerated reaction rates, improved yields, and retained activity across multiple cycles, highlighting its efficiency and reusability. These results demonstrate not only a practical and sustainable route for heterocyclic compound synthesis but also contribute to the broader field of nanocatalysis by showcasing how CoFe_2O_4 nanoparticles can enable greener chemical processes. This work advances existing knowledge by integrating nanotechnology with organic synthesis, offering a scalable approach that aligns with SDG-12: Responsible Consumption and Production through reduced waste, energy efficiency and catalyst recyclability. Future studies may extend this methodology to industrial-scale applications and explore biomedical relevance, thereby strengthening the role of nanocatalysts in sustainable innovation.

ACKNOWLEDGMENTS

All Authors are thankful to the Principal, Dr Rafiq Zakaria College For Women, Chhatrapati Sambhajinagar-431001, (Maharashtra), India, for providing us with the facility of the research Laboratory.

CONFLICT OF INTERESTS

The Authors declare that they have no conflict of interest.

AUTHOR CONTRIBUTIONS

The present work is related to *SDG-12 Responsible Consumption and Production*. All the authors contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

Komal V. Takankhar  <https://orcid.org/0009-0007-6847-4201>

Ayesha N. Durrani  <https://orcid.org/0009-0000-8384-2446>

Open Access: This article is distributed under the terms of the Creative Commons Attribution 4.0 International License (<http://creativecommons.org/licenses/by/4.0/>), which permits unrestricted use, distribution, and reproduction in any medium, provided you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

Cite this article as: Komal V. Takankhar and A. Durrani, *Rasayan J. Chem.*, 19(4), 1961-1966(2026), <https://doi.org/10.31788/RJC.2026.1949869>

REFERENCES

1. R. Arulmurugan, G. Vaidyanathan, S. Sendhilnathan and B. Jeyadevan, *Journal of Magnetism and Magnetic Materials*, **298**, 83(2006), <https://doi.org/10.1016/j.jmmm.2005.03.002>
2. E.V.Gopalan, I.A. Al-Omari, D.S. Kumar, Y. Yoshida, P. A. Joy, and M.R. Anantharaman, *Applied Physics A*, **99**, 497(2010), <https://doi.org/10.1007/s00339-010-5573-8>
3. P. Kidkhunthod, S. Nilmoung, S.Mahakot, S. Rodporn, S.Phumying, and S. Maensiri, *Journal of Magnetism and Magnetic Materials*, **401**, 436(2016), <https://doi.org/10.1016/j.jmmm.2015.10.085>
4. S. Mirzaee, S. F. Shayesteh and S. MahdaviFar, *Polymer*, **55**, 3713(2014), <https://doi.org/10.1016/j.polymer.2014.06.039>
5. R. Topkaya, U. Kurtan, Y. Junejo and A. Baykal, *Materials Research Bulletin*, **48**, 3247(2013), <https://doi.org/10.1016/j.materresbull.2013.04.071>
6. Xu HJ, X. Wan, Y. Geng, Xu XL. *Current Organic Chemistry*, **17**, 1034(2013), <https://doi.org/10.2174/1385272811317100006>
7. C. W. Lim, and I. S. Lee, *Nano Today*, **5**, 412(2010), <https://doi.org/10.1016/j.nantod.2010.08.008>
8. P. Jehani, R Siddique, D.Gautam, M. Panchal, P. Gautam, and R. K. Bera, *Journal of Heterocyclic Chemistry*, **63**, 648(2026) <https://doi.org/10.1002/jhet.70131>
9. D. Kaminsky, A. Kryshchyshyn, and R. Lesyk, *European Journal of Medicinal Chemistry*, **140**, 542(2017), <https://doi.org/10.1016/j.ejmech.2017.09.031>
10. M. T. Chhabria, S. Patel, P. Modi, and P. S. Brahmshatriya, *Current Topics in Medicinal Chemistry*, **16**, 2841(2016), <https://doi.org/10.2174/1568026616666160506130731>
11. M. A.Gouda, M. A. Berghot, G. E. Abd El-Ghani, and A. M. Khalil, *European Journal of Medicinal Chemistry*, **45**, 1338(2010), <https://doi.org/10.1016/j.ejmech.2009.12.020>

[RJC-9869/2026]