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An Efficient Strategy for Quinazolinone and Benzimidazole Synthesis Using Microwave-Assisted Copper-Catalyzed Aerobic Oxidation of Amines

Shyamal Kanti Bera,[a] Muhammad Irfan,[a] and Andrea Porcheddu*[a]

Microwave-assisted organic synthesis (MAOS) has emerged as an effective technique for accelerating chemical reactions while enhancing efficiency and selectivity. In this study, we developed a copper-catalyzed method for aerobic oxidative coupling to synthesize quinazolinones and benzimidazoles, using oxone as the oxidant under microwave irradiation. This method successfully couples amines with various partners, including 2-aminobenzamides and *o*-phenylenediamines. The strategy demonstrates broad substrate compatibility, exhibits good functional group tolerance, and consistently achieves moderate yields, making it a versatile and efficient approach for synthesizing heterocycles.

1. Introduction

Nitrogen-containing heterocyclic compounds,[1] particularly quinazolinones[2] and benzimidazoles,[3] are pivotal scaffolds in pharmaceutical development due to their diverse biological activities[4] and broad therapeutic applications.[5] Quinazolinones, distinguished by their bicyclic nitrogenous structure, are gaining attention for their potential in anticancer,[6] antimicrobial,[7] and anti-inflammatory therapies,[8] underscoring their critical role in modern medicine.[9] Likewise, benzimidazoles[10] exhibit notable antiviral,[11] antifungal,[12] and antiparasitic properties,[13] establishing them as essential components in medicinal chemistry.[14] Furthermore, 1,2-disubstituted benzimidazoles act as effective antiviral agents and selective receptor antagonists and serve as valuable intermediates in dye and polymer synthesis.[15] The ability of these compounds to serve as scaffolds for bioactive molecules underscores their therapeutic potential in tackling global health issues.[16] As the demand for innovative treatments increases, effective synthetic strategies for quinazolinones and benzimidazoles become essential as their distinct structural motifs and varied bioactivities continue to drive advancements in pharmaceutical science.

Various synthetic methods have been developed in recent years to prepare quinazolinones, with a few notable examples illustrated in Scheme 1. Among these, the most commonly used strategy involves the application of 2-aminobenzamide[17] and its derivatives as crucial starting materials, typically reacting with aldehydes or similar compounds[18] to produce a variety of quinazolinone derivatives under both metal-catalyzed[19] and metal-free conditions.[20] Several metal-catalyzed reactions (employing Pd,[21] Pt,[22] Ir,[23] Fe,[24] Ru,[25] and Mn[26]) have been extensively reported in the literature to synthesize quinazolinones. Traditional approaches often require high temperatures, long reaction times, costly metal catalysts, harsh conditions, or multiple steps. The efficiency of these methods is somewhat limited when considering atom economy. Consequently, there is an increasing demand for a one-step, atom-economical approach to synthesize quinazolinones and benzimidazoles directly from readily available starting materials.

Microwave-assisted synthesis[27] has significantly advanced the field of organic chemistry[28] by providing a more efficient and sustainable[29] alternative to traditional heating methods,[30] particularly in synthesizing bioactive heterocycles[31] like benzimidazoles[32] and quinazolinones.[33] Unlike classical methods that depend on slow external heat transfer, microwave irradiation offers rapid, uniform heating at the molecular level. This significantly accelerates reaction rates and enhances the efficiency of copper-catalyzed oxidative coupling reactions.[34] This leads to quicker reaction times, better yields, and improved selectivity while reducing by-products. Moreover, the mild conditions provided by microwave irradiation lessen the need for harsh reagents, making the process more environmentally friendly and energy efficient. The ability to carry out these transformations under greener conditions supports more sustainable practices in synthetic organic chemistry,[35] making microwave-assisted copper-catalyzed synthesis a powerful tool for developing necessary critical nitrogen-containing scaffolds with significant biological activity.

The oxidation of amines,[36] accompanied by carbon-heteroatom bond formation,[37] is recognized as a crucial transformation in synthetic organic chemistry, given its extensive range of applications across various domains.[38] Using benzylamine in organic synthesis is an essential conduit for

[a] S. K. Bera, M. Irfan, A. Porcheddu
Dipartimento di Scienze Chimiche e Geologiche, Università degli Studi di Cagliari, Cittadella Universitaria, Cagliari 09042, Italy
E-mail: porcheddu@unica.it

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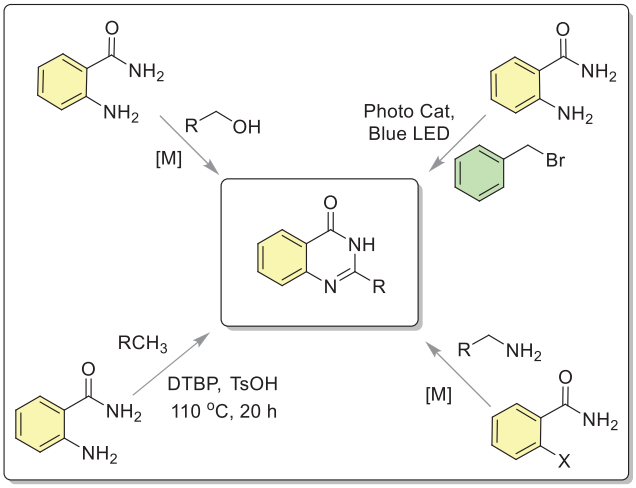
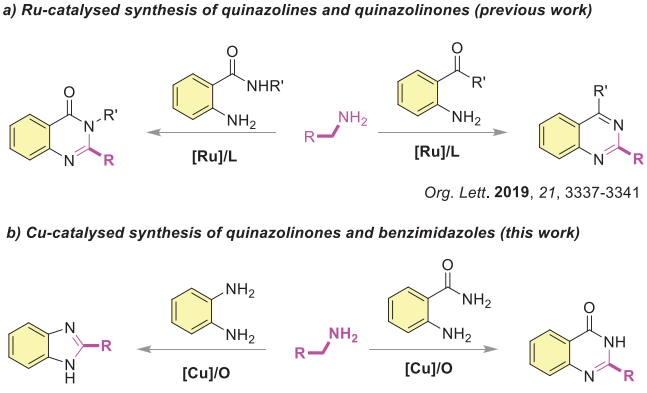


Scheme 1. Various synthetic methods for producing quinazolinones derivatives from different starting materials.



Scheme 2. Utilization of amines. (a) Previous work: Ru-catalyzed synthesis of quinazolines and quinazolinones. (b) This work: Copper-catalyzed synthesis of quinazolinones and benzimidazoles.

Table 1. Optimization of the reaction conditions.[a]

Entry	Cat. (mol%)	Ox. (Equiv)	Solvent	Yield[b]
1	$Cu(OAc)_2$ (10)	Oxone (1)	DMSO	64%
2	$Cu(OAc)_2$ (10)	Oxone (1)	DMF	60%
3	**$Cu(OAc)_2$ (10)**	**Oxone (1)**	**PhMe**	**89%**
4	$Cu(OAc)_2$ (10)	Oxone (0.75)	PhMe	69%
5	$Cu(OAc)_2$ (10)	Oxone (1.25)	PhMe	89%
6	$Cu(OAc)_2$ (5)	Oxone (1)	PhMe	75%
7	I_2 (10)	Oxone (1)	PhMe	65%
8	CuI (10)	Oxone (1)	PhMe	69%
9	$Cu(OTf)_2$ (10)	Oxone (1)	PhMe	86%
10	$CuSO_4{\cdot}5H_2O$ (10)	Oxone (1)	PhMe	64%
11	$Cu(OAc)_2$ (10)	Oxone (1)	H_2O	82%
12[c]	$Cu(OAc)_2$ (10)	Oxone (1)	EtOH	35%
13	$Cu(OAc)_2$ (10)	Urea·H_2O_2 (1)	PhMe	59%
14	$Cu(OAc)_2$ (10)	$K_2S_2O_8$ (1)	PhMe	71%
15[c]	$Cu(OAc)_2$ (10)	Oxone (1)	PhMe	68%
16[d]	$Cu(OAc)_2$ (10)	Oxone (1)	PhMe	76%
17[e]	$Cu(OAc)_2$ (10)	Oxone (1)	PhMe	67%
18	$Cu(OAc)_2$ (10)	Oxone (1)	–	0%
19[f]	$Cu(OAc)_2$ (10)	Oxone (1)	H_2O	27%
20	$Cu(OAc)_2$ (10)	Oxone (1)	CPME[g]	36%
21	$Cu(OAc)_2$ (10)	Oxone (1)	Ionic liquid[h]	41%

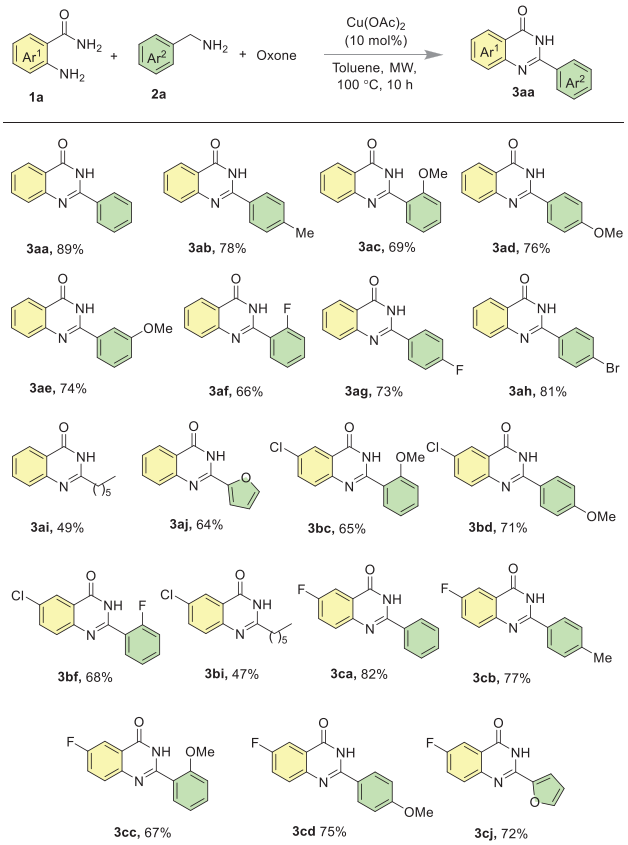
[a] Reaction condition: All the reactions were performed using 0.5 mmol of anthranilamide (**1a**) and 0.5 mmol of benzyl amines (**2a**) in 2 mL of solvent at 100 °C stirring for 10 h.
[b] Isolated yield.
[c] Reaction temperature 80 °C.
[d] Reaction temperature 120 °C.
[e] Reaction time 8 h.
[f] Sodium dodecyl sulfate (SDS) was used as a surfactant in water.
[g] CPME (cyclopropyl methyl ether).
[h] Ionic liquid: (1-decyl-3-methyl-imidazolium tetrafluoroborate).

forming diverse heterocycles,[39] which are esteemed for their wide-ranging pharmacological properties.[40] This reaction is not just a synthetic step but a crucial strategy that unlocks the potential for developing innovative therapeutic agents. The ability to efficiently convert benzylamine into these complex heterocycles underscores its significance in medicinal chemistry,[41] as it paves the way for discovering novel drugs that can address many diseases.

In this context, Yi and colleagues elucidated an innovative strategy employing ruthenium-catalyzed dehydrogenative and deaminative coupling of amines, paving the way for synthesizing intricate quinazoline and quinazolinone derivatives (Scheme 2a).[42] By leveraging advanced oxidative methodologies, researchers can enhance the accessibility and diversity of quinazolinones and benzimidazoles, thereby propelling the advancement of pharmaceutical science. This study presents an elegant and efficient method for the copper-catalyzed aerobic oxidative coupling of various benzylamines with 2-aminobenzamide and *o*-phenylenediamine. This process leads to the formation of quinazolinones and benzimidazole heterocycles. Driven by microwave irradiation, the method showcases remarkable simplicity and significantly reduced reaction times, highlighting its potential as an advanced approach to synthesizing heterocycles.

2. Results and Discussion

Anthranilamide (**1a**) and benzylamine (**2a**) were selected as model substrates to optimize the reaction conditions (Table 1). In the initial experiment, using 10 mol% of $Cu(OAc)_2$ as the catalyst and one equivalent of Oxone in DMSO as the solvent produced 2-phenylquinazolin-4(3*H*)-one (**3aa**) with a yield of 64% after 10 h of microwave irradiation at 100 °C. A comparable yield was achieved using DMF. However, switching to toluene significantly enhanced the yield to 89% (entry 3, Table 1). Reducing the quantity of Oxone to 0.75 equivalents resulted in a lower yield of 69%. However, increasing it to 1.25 equivalents under the same conditions did not improve the outcome. Using iodine instead of copper acetate as the catalyst yielded a 65% yield (entry 7, Table 1). Other copper salts such as CuI,



Scheme 3. Synthesis of quinazolinones from various benzyl amines.

$Cu(OTf)_2$, and $CuSO_4{\cdot}5H_2O$ were tested; however, $Cu(OAc)_2$ was the most efficient for this aerobic oxidative coupling. Interestingly, using water as a solvent resulted in an 82% yield; however, it required an additional workup process for further purification. In contrast, using ethanol as the solvent drastically reduced the yield to 35%. Alternative oxidants like urea hydrogen peroxide and $K_2S_2O_8$ were also evaluated but failed to enhance the results. Neither raising nor lowering the reaction temperature from 100 °C improved the yield, and shortening the reaction time yielded inferior results (entry 17, Table 1). However, no product formation was observed when the reaction was performed under neat conditions (solvent-free) (entry 18). Using sodium dodecyl sulfate (SDS) as a surfactant in water under the optimized reaction conditions, the yield of **3aa** dropped significantly from 89% to 27% (entry 19). Moreover, replacing toluene with the green solvent CPME (cyclopropyl methyl ether) resulted in a substantial decrease in yield to 36% (entry 20). Similarly, employing the green solvent ionic liquid (1-Decyl-3-methylimidazolium tetrafluoroborate) caused the yield to drop significantly to 41% (entry 21). Ultimately, the optimal conditions were established with 10 mol% of $Cu(OAc)_2$ and 1 equivalent of Oxone in toluene at 100 °C for 10 h under microwave irradiation, providing 2-phenylquinazolin-4(3*H*)-one (**3aa**) in 89% yield (entry 3, Table 1).

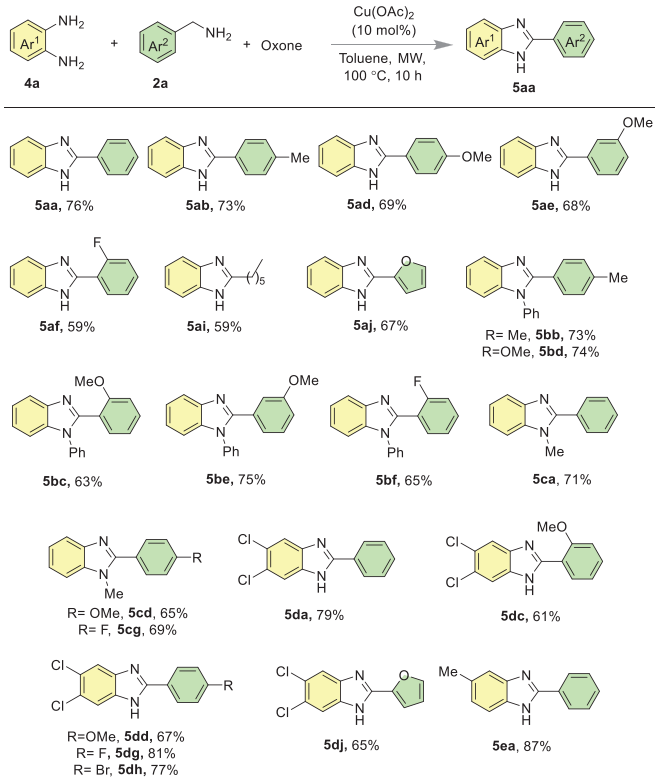
With the optimized reaction conditions established, we thoroughly explored the substrate versatility of the Cu-catalyzed aerobic oxidative coupling process, utilizing a wide range of substituted amines and anthranilamides, as shown in Scheme 3. Various electron-donating and electron-withdrawing amines demonstrated excellent compatibility with the reaction conditions, reliably delivering the desired products in substantial yields.

Unsubstituted anthranilamide, for instance, reacted efficiently with para-methyl benzylamine to furnish product **3ab** in an impressive 78% yield. Benzylamines with methoxy groups afforded yields between 69% and 76%, while fluoro-substituted benzylamines smoothly transformed into **3af** and **3ag** with respectable yields of 66% and 73%, respectively. Notably, benzylamine bearing a bromo group was proficiently converted to product **3ah**, achieving a remarkable 81% yield. Heptylamine and furfurylamine reacted effectively under standard conditions, yielding their respective products in 49% and 64% yields. Furthermore, methoxy and fluoro-substituted benzylamines coupled with chloro-substituted anthranilamide generate products **3bc**, **3bd**, and **3bf** in good yields. The aliphatic amine, heptylamine, also cyclized efficiently, affording product **3bi** with a 47% yield. Additionally, unsubstituted benzylamines reacted effortlessly with fluoro-substituted anthranilamides to deliver product **3ca** in an impressive 82% yield.

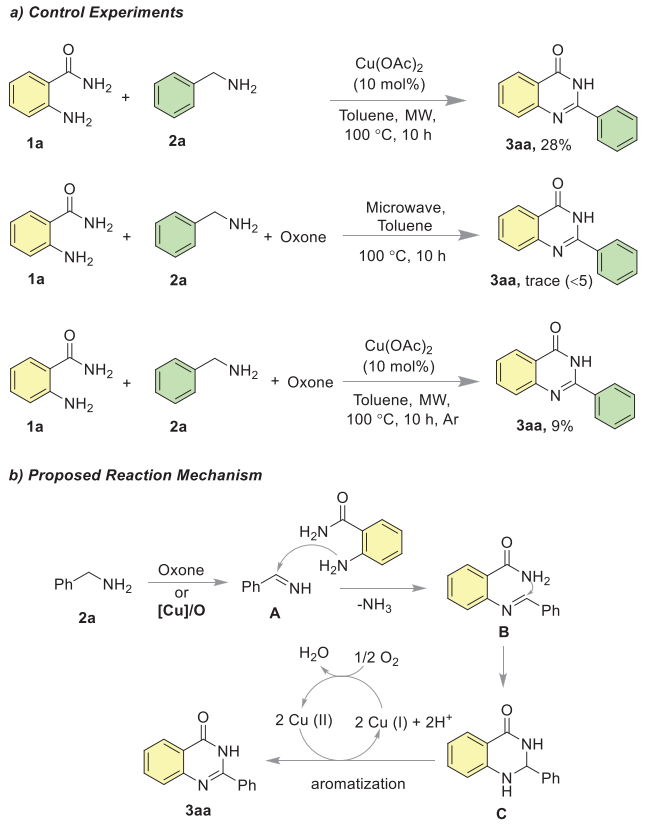
Methyl- and methoxy-substituted benzylamines also efficiently coupled with fluoro-substituted anthranilamides, yielding products **3cb**–**3cd** in moderate yields. Remarkably, furfuryl amine was well-tolerated under standard conditions, affording product **3cj** with a 72% yield. In this regard, we have performed the reaction using sterically hindered benzylamine (1,1 diphenylmethylamine) under the standard reaction condition. We observed the cyclized product around 2%, which was confirmed by GCMS. We are unable to isolate the product. The GCMS data are attached in the Supporting Information.

To explore the diversity of substrates in the Cu-catalyzed aerobic oxidative coupling method, we conducted a detailed study of various amines under optimized reaction conditions, using *o*-phenylenediamine derivatives as coupling partners. The outcomes of this investigation are elegantly depicted in Scheme 4. Under the standard reaction conditions, benzylamine was reacted with *o*-phenylenediamine, forming 2-phenyl-1*H*-benzo[d]imidazole (**5aa**) with a yield of 76%. Introducing methyl, methoxy, and fluoro groups into the benzylamine structure produced products (**5ab**–**5af**) with good yields. Similarly, using heptylamine led to the formation of product **5ai**, which yielded 59%. Notably, furfurylamine produced the corresponding compound (**5aj**) with a 67% yield. Interestingly, the presence of substituents on the nitrogen of *o*-phenylenediamine, such as phenyl and methyl, was well-tolerated, delivering the desired products with moderate yields. In this context, *N*-phenyl-*o*-phenylenediamine reacted with benzylamines containing methyl, methoxy, and fluoro groups, yielding the respective products (**5bb**–**5bf**) in good yields. The reactivity of *N*-methyl-*o*-phenylenediamine was also excellent toward benzylamine, as well as methoxy- and fluoro-substituted benzylamines, producing products **5ca** and **5cd** and **5cg** with yields of 71%, 65%, and 69%, respectively. Chloro-substituted *o*-phenylenediamine proved well-tolerated, providing **5da** with a 79% yield.

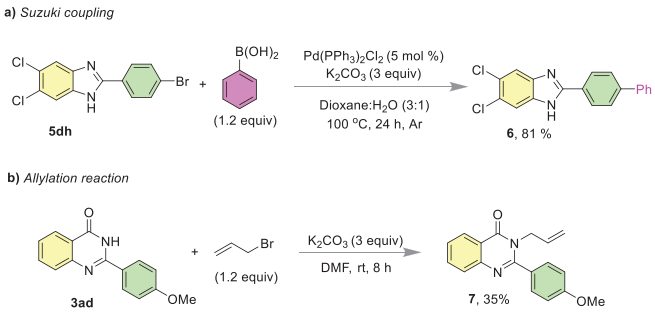
Benzylamines substituted with methoxy, fluoro, and bromo groups reacted smoothly with 4,5-dichlorobenzene-1,2-diamine, delivering the corresponding products (**5dc**, **5dd**, **5dg**, and **5dh**)



Scheme 4. Synthesis of benzimidazoles from various amines.



Scheme 5. (a). Control experiments. (b) Proposed reaction mechanism.



Scheme 6. Synthetic utilities of the synthesized compound. (a) Suzuki coupling reaction of **5dh**. (b) Allylation of **3ad**.

in yields ranging from 61% to 81%. Notably, furfurylamine also delivered **5dj** with a 65% yield. Additionally, 4-methylbenzene-1,2-diamine actively participated under the standard reaction conditions, affording product **5ea** with an impressive 87% yield.

Some control experiments were carried out to shed light on the reaction mechanism, as depicted in Scheme 5a. Initially, the reaction was carried out without including an external oxidant (oxone), yielding the desired product at 28%. This observation indicates that the copper catalyst alone cannot significantly enhance product yield, highlighting the need for an external oxidant. Interestingly, when the reaction occurred without the copper catalyst, oxone alone produced only a trace amount of the product (Scheme 5a). This suggests neither the copper catalyst nor oxone alone is sufficient for optimal conversion.

In contrast, the synergistic combination of the copper catalyst and oxone under aerobic conditions delivered the product with an impressive yield of 89% (Table 1, entry 3). When the reaction was performed without copper and under an argon atmosphere, the product yield dropped significantly to only 9% (Scheme 5a). This underscores the essential roles of both the copper catalyst and molecular oxygen in the reaction's success.

We propose the reaction mechanism outlined in Scheme 5b based on our control experiments and existing literature.[43] The process begins with the oxidation of benzylamine, which forms phenylmethanimine (**A**) through oxone treatment under aerobic conditions, likely involving the formation of hydroxylamine intermediates.[44] Next, the amine group of anthranilamide performs a nucleophilic attack on intermediate **A**, forming intermediate **B**. This intermediate then undergoes intramolecular cyclization to produce intermediate **C**. Finally, intermediate **C** goes through oxidative dehydrogenation,[45] ultimately forming the desired product, **3aa**.

The synthetic applications of our synthesized compounds (**5dh** and **3ad**) are illustrated in Scheme 6. First, a Suzuki coupling reaction between **5dh** and phenylboronic acid produced 2-([1,1′-biphenyl]-4-yl)-5,6-dichloro-1*H*-benzo[d]imidazole (compound **6**) with a yield of 81%. Next, we explored the N-H allylation of compound **3ad** using allyl bromide. When 1.2 equivalents of allyl bromide and 3 equivalents of K_2CO_3 were used in DMF at room temperature, the reaction afforded our expected *N*-alkylated product (**7**) in 35% yield along with the formation of *O*-alkylated product with 65% yield.

3. Conclusion

In conclusion, we have developed a copper-catalyzed aerobic oxidative methodology for synthesizing quinazolinones and benzimidazoles, representing a significant advancement in heterocyclic chemistry. This approach offers an efficient, cost-effective, and environmentally friendly alternative to traditional methods by utilizing readily available starting materials. Demonstrating a broad substrate scope with good yields, it underscores both versatility and practicality. The microwave-assisted modification further enhances the process by reducing reaction times and temperatures, aligning with green chemistry principles. This method simplifies the synthesis of nitrogen-containing heterocycles and holds considerable promise for various applications in pharmaceutical and materials sciences. Consequently, it positions itself as a valuable tool for future innovations in synthetic chemistry. Our ongoing studies are focused on exploring additional applications of this cascade method for constructing nitrogen-containing heterocycles.

Supporting Information

The detailed experimental procedure and the ^{1}H and ^{13}C NMR analysis data are provided in the Supporting Information.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the Supporting Information of this article.

Keywords: Aerobic oxidation · Benzimidazoles · Copper catalyst · Microwave irradiation · Quinazolinones

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