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[3 + 2] Cycloaddition of Nitrile Imines with Enamides: An Approach to Functionalized Pyrazolines and Pyrazoles

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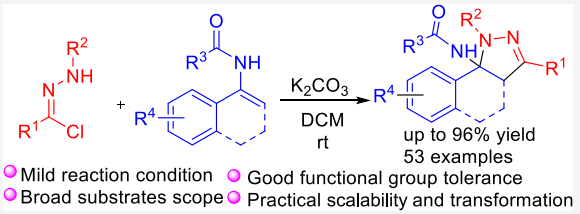
ABSTRACT: An efficient [3 + 2] cycloaddition of in situ generated nitrile imines with enamides has been established. A wide range of functionalized pyrazoline derivatives (53 examples) were obtained in moderate to good yields (up to 96%) under very mild conditions. This protocol features broad substrate scope, good functional group tolerance, and operational simplicity. Practical transformation of the products into useful pyrazoles via a one-pot process and the scalability of this protocol highlight the utility of this synthetic methodology.



INTRODUCTION

Nitrogen-containing heterocyclic compounds represent versatile structural motifs in natural products, pharmaceuticals, and functional materials.[1] In particular, pyrazoline and pyrazole skeletons have received enormous attention as a ubiquitous structural unit in pesticides and pharmaceutical compounds displaying a wide range of biological activities (Figure 1).[2] For instance, compound I showed antitubercular activity.[3] Compound II was reported to be a CB-receptor ligand.[4] Apixaban, which contains a pyrazole core structure, has proved to be a factor Xa inhibitor for the treatment of cardiovascular diseases.[5] Thus, development of efficient and versatile synthetic strategies for the preparation of these moieties has received enormous research interest, and various elegant methods have been reported (Scheme 1). Traditional access to pyrazole and pyrazoline derivatives mainly involves condensation of hydrazines with dicarbonyl compounds or unsaturated carbonyls (Scheme 1a).[6] During the past decade, cycloadditions of two-nitrogen synthons with various partners have been established as the most prominent strategies for the synthesis of pyrazolines. For example, [3 + 2] cycloaddition reactions of Bestmann−Ohira reagent (BOR) and Seyferth−Gilbert reagent (SGR) with olefinic derivatives to access spiro-phosphonylpyrazolines have been developed by Peng and Mohanan (Scheme 1b).[7] In addition, [4 + 1] annulation reactions of in situ formed azoalkenes with C1 synthons were also successfully demonstrated for the construction of pyrazolines by several groups (Scheme 1c).[8] Recently, [3 + 2] cycloadditions of nitrile imines with alkenes represent an attractive strategy to prepare pyrazolines (Scheme 1d).[9] For example, Feng and Stanley developed asymmetric 1,3-dipolar cycloaddition reactions of nitrile imines with 3-alkenyl-oxindoles to afford chiral spiro-pyrazoline-oxindoles, respectively.[10] Guo reported 1,3-dipolar cycloaddition of nitrilimines with allenoates to afford spiro-bidihydropyrazoles.[11] Su described [3 + 2] cycloaddition of nitrile imines with *para*-quinone methides to synthesize spiro-pyrazoline-cyclohexadienones.[12] Despite these elegant strategies, development of a novel method to generate pyrazolines remains highly desirable.

On the other hand, although the cycloadditions of nitrile imines have been well developed in the past decades, the most reported strategies involving cycloaddition of nitrile imines are mainly limited to the electron-deficient olefinic partners[9−13] except for a few examples,[14] which largely limits application of this strategy. Therefore, development of a novel method to construct pyrazolines using nitrile imine with electron-rich C═C derivatives is challenging and highly desirable, which would expand the scope of nitrile imines and access pyrazolines that cannot be obtained by other methods.

Enamides are versatile building blocks in organic synthesis and have been widely employed as substrates in various functionalization reactions[15] and cycloadditions to construct nitrogen-containing compounds.[16] However, to our knowledge, cycloaddition reaction of enamides with nitrile imines has not been reported. We hypothesized that the nucleophilic and electrophilic enamides would be matched with nitrile imines for the preparation of pyrazolines via [3 + 2] cycloaddition. Additionally, biologically important pyrazoles would also be efficiently obtained by subsequent elimination of the acyl group. We herein report the first [3 + 2] cycloaddition reaction

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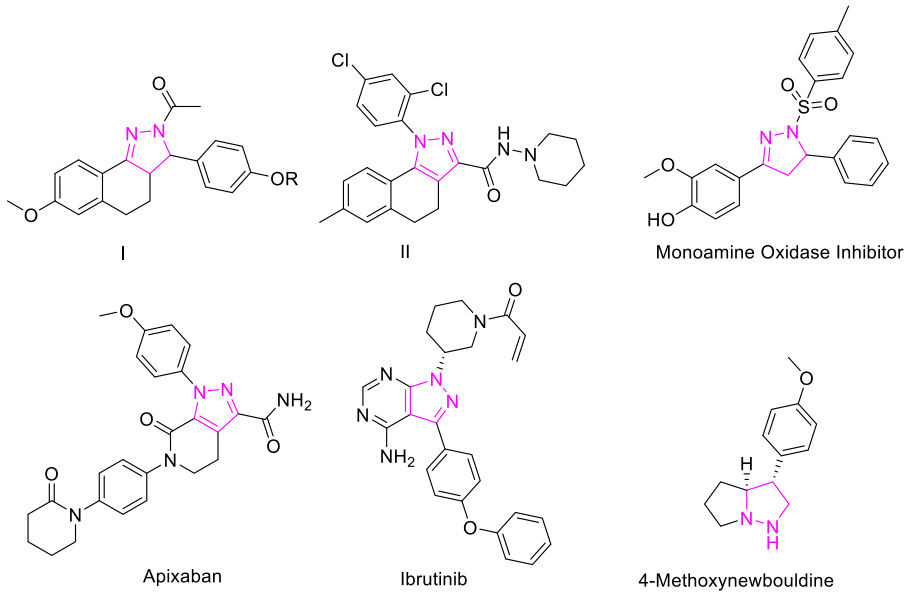
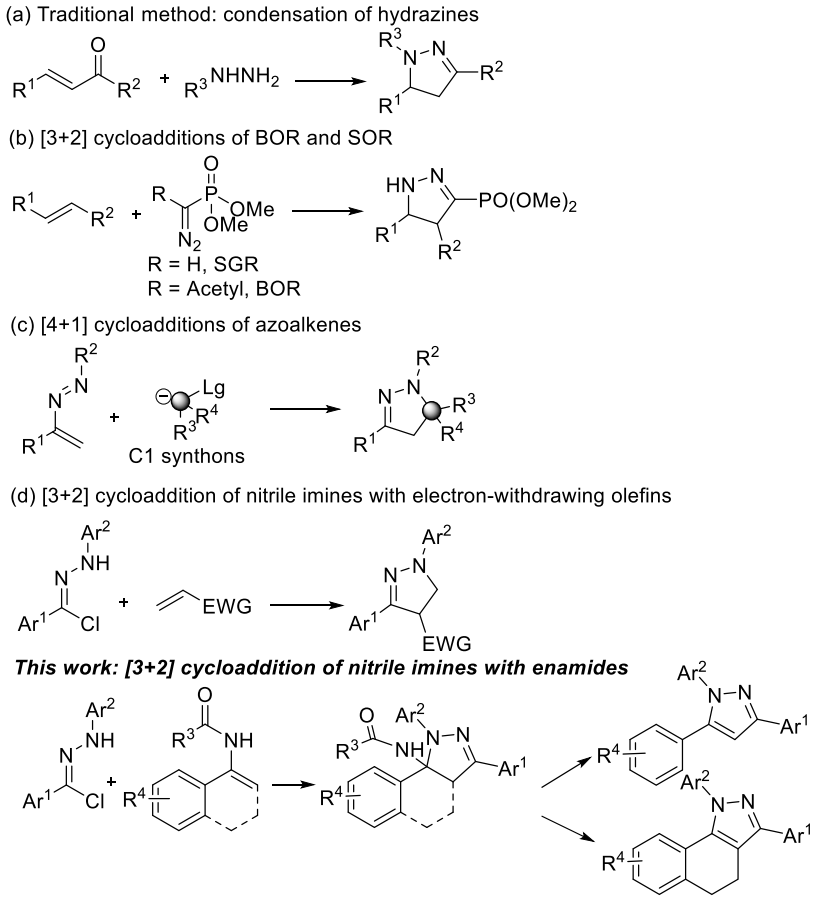


Figure 1. Representive drugs and biologically active compounds bearing pyrazoline and pyrazole units.

Scheme 1. Different Strategies for Pyrazolines Synthesis



between in situ generated nitrile imines and enamides for the efficient synthesis of pyrazolines.

RESULTS AND DISCUSSION

We began our study using the readily accessible *N*-phenylbenzohydrazonoyl chloride **1a** and *N*-(3,4-dihydronaphthalen-1-yl)acetamide **2a** as the model substrates to test the feasibility of the designed [3 + 2] cycloaddition reaction (Table 1). With the use of Na_2CO_3 as the base, the reaction was performed in 1,2-dichloroethane at room temperature for 40 h to give the desired product fused pyrazoline **3aa** as a single diastereoisomer in 66% yield (Table 1, entry 1). Investigation on solvents indicated that CH_2Cl_2 and $CHCl_3$ are also suitable for the reaction (entries 2 and 3). It was found that the oxygenated

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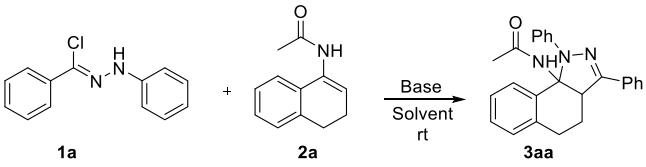
Table 1. Optimization of Reaction Conditions[a]



1a 2a 3aa

entry	Base	solvent	t (h)	yield[b]
1	Na_2CO_3	DCE	40	66
2	Na_2CO_3	DCM	24	72
3	Na_2CO_3	$CHCl_3$	40	62
4	Na_2CO_3	dioxane	72	40
5	Na_2CO_3	THF	72	37
6	Na_2CO_3	MeOH	72	46
7	Na_2CO_3	MeCN	60	79
8	Na_2CO_3	toluene	40	53
9	Na_2CO_3	DMF	5	65
10	$NaHCO_3$	DCM	72	27
11	NaOH	DCM	72	42
12	DIPEA	DCM	72	59
13	TEA	DCM	72	53
14	DABCO	DCM	72	45
15	K_2CO_3	DCM	24	92
16[c]	K_2CO_3	DCM	48	85
17[d]	K_2CO_3	DCM	24	92
18[e]	K_2CO_3	DCM	36	75

[a]The reactions were performed with **1a** (0.3 mmol), **2a** (0.2 mmol), and base (0.6 mmol) in the solvent (2.0 mL) at room temperature. [b]Isolated yields, only one diastereoisomer was observed by crude NMR analysis. [c]0.4 mmol K_2CO_3 was used. [d]**1a** (0.4 mmol), **2a** (0.2 mmol). [e]**1a** (0.22 mmol), **2a** (0.2 mmol).

solvent has a detrimental effect on the reaction for an unknown reason, giving low to moderate yields even with prolonged time (entries 4−6). Further screening of the solvents resulted in no improvement of the reaction efficiency (entries 7−9). It is worth noting that when the reaction was exposed in dimethylformamide (DMF), the reaction time was decreased to 5 h from the previous 24 h, although the yield of **3aa** decreased slightly (entry 9). Then, different bases were screened to improve the yield (entries 10−15). Unfortunately, $NaHCO_3$, NaOH, or organic bases only gave moderate yields (entries 10−14). To our delight, the yield was increased to 92% with the use of K_2CO_3 as the base. Lowering the amount of the base resulted in a slight decrease of the yield (entries 15 and 16). Attempts on further improving the efficiency of the reaction by screening the ratios of the substrates were unsuccessful (entries 17 and 18). As a result, a combination of K_2CO_3 (3.0 equiv) and CH_2Cl_2 at room temperature was established as the most suitable reaction condition.

With the optimized reaction conditions in hand, we emphasize on exploring the substrate scope of this protocol by employing a variety of hydrazonyl chlorides **1**. Initially, hydrazonyl chlorides derived from various substituted phenylhydrazines and benzoyl chloride were examined, and the results are summarized in Scheme 2. Hydrazonyl chlorides **1** with electron-donating groups, including *ortho*-substituted hydrazonyl chlorides, were good substrates and gave the desired pyrazolines in good to excellent yields (87−96%) (Scheme 2, **3aa**−**ca**, **3fa**, **3ma**). However, electron-withdrawing substituents have an adverse effect on reaction efficiency. Hydrazonyl chlorides with electron-withdrawing groups resulted in slightly lower yields compared to the electron-donating ones (Scheme 2, **3da**, **3ea** vs **3aa**, **3ba**). Although the substrate bearing electron-donating *ortho*-substituent gave decent yield (**3fa**), those with electron-withdrawing *ortho*-substituted halogen led to low to moderate yields (**3ga**, **3ha**, **3ia**, **3ja**). These results showed that electron-donating substituents on the phenyl ring may increase the electron cloud density of the nitrogen, which is beneficial to the reaction. The significant steric effect of the substituent on the aromatic ring of hydrazonoyl chlorides on the reaction efficiency has also been observed (**3da**, **3ea**, **3ga**). Then, the effect of substituents linked to C═N was investigated. Generally, substrates with electron-donating or electron-withdrawing group on the phenyl ring afforded the products in 80−90% yield. Ester group was also tolerated in these reaction conditions and delivered the desired product **3ta** in 87% yield. Moreover, substrates with heteroaryl, such as furanyl and thienyl, generated the corresponding products in uncompromising yields (**3ua** and **3va**). Notably, the aliphatic-substituted hydrazonyl chloride could also be subjected to the reaction. However, the product was very unstable and decomposed to the corresponding pyrazole in chloroform even without any promotor (**3wa**).

To further examine the generality of the reaction, we next investigated the scope of different cyclic enamindes under the established conditions to prepare the fused pyrazolines. First, we tested a range of benzo five-membered cyclic enamides bearing various substituents on the benzene ring. Electron-rich enamides showed good reactivity to yield the product in excellent yields (Scheme 3, **3ab**, **3ac**). Enamides with electron-withdrawing substituents led to relatively lower yields (Scheme 3, **3ad**, **3ae**, **3af**). A ring expansion as in enamides (n = 2, 3) was possible without any difficulty and gave the product in 82 and 73% yields, respectively (Scheme 3, **3ag**, **3ah**). Then, different *N*-benzoyl enamides were investigated. In general, the transformation displayed high functional group tolerance and proved to be a reliable methodology for the preparation of multisubstituted pyrazolines in 74−89% yields (Scheme 3, **3ai**−**as**). However, *N*-naphthoyl enamide led to fair yield, probably due to the steric hindrance (**3at**). Finally, we tried to expand the substrate scope to aliphatic-substituted enamides, and excellent yields were obtained (**3au**−**aw**). It is worth noting that this reaction proceeded in a highly diastereoselective manner and also only one diastereoisomer was observed by crude NMR (**3aw**).

Based on the above examples, we envisioned that if acyclic enamides were used to couple with nitrile imines, the biologically important and highly functionalized nonfused pyrazolines would be obtained. As shown in Scheme 4, a series of acyclic enamides with electron-withdrawing or electron-donating substituents on the benzene ring could smoothly perform the reaction under mild conditions, affording the desired pyrazolines in good to excellent yields (82−90%) (Scheme 4, **4a**−**i**).

To further demonstrate the practical utility of this method, a one-pot synthesis of pyrazoles and tetralone-fused pyrazoles was conducted via [3 + 2] cycloaddition reaction, followed by subsequent elimination induced by a catalytic amount of $AlCl_3$. A wide range of acyclic and cyclic *N*-acetyl enamides were reacted with nitrile imines to synthesize the useful pyrazole derivatives (Scheme 5, **5a**−**q**). The acid-sensitive methoxyl and *tert*-butyl on the benzene ring of enamides are also compatible with the reaction conditions, giving the product in 91 and 87% yields (Scheme 5, **5g** and **5h**), respectively. The ester-substituted hydrazonyl chloride performed the reaction to yield the product **5n** in 87% yield.

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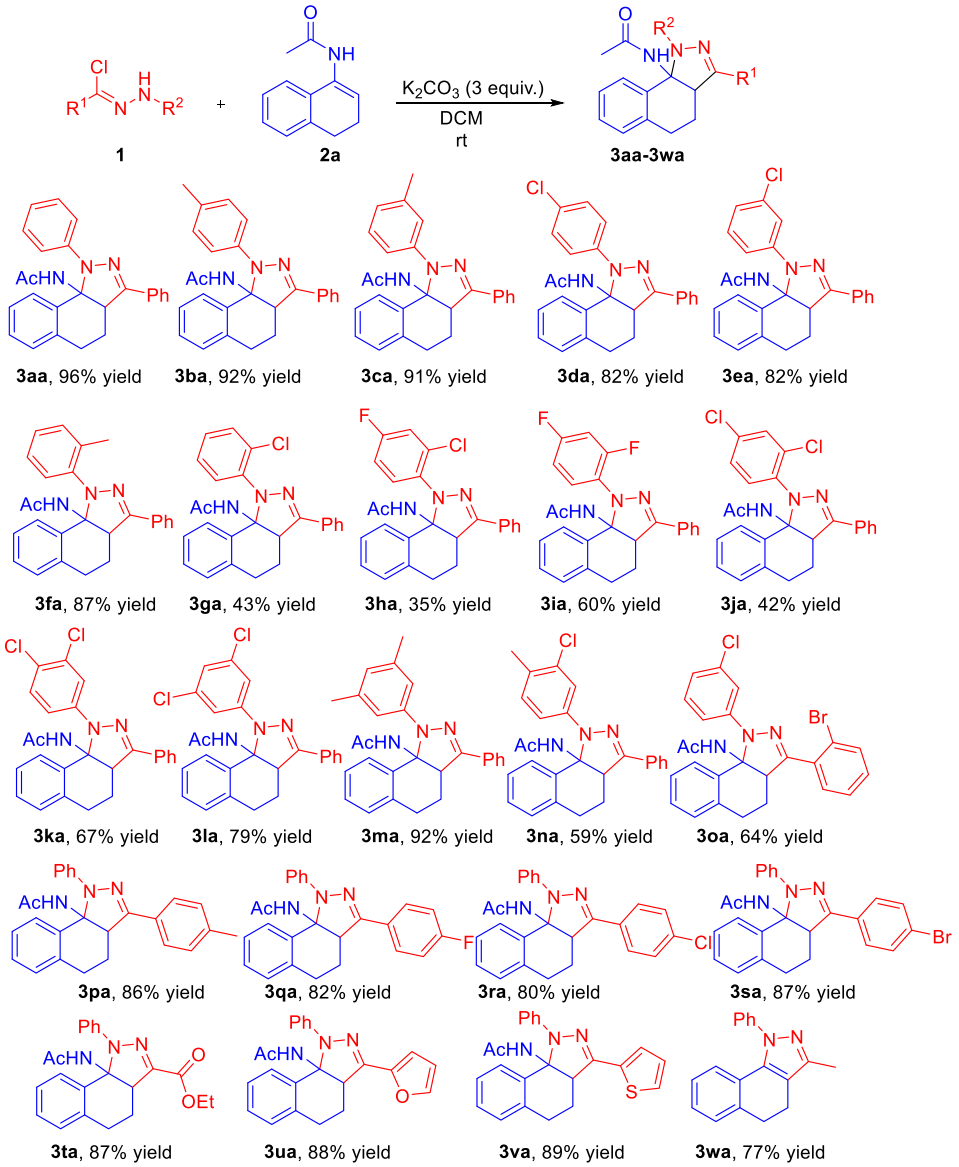
Scheme 2. Substrate Scope of Hydrazonoyl Chloride[a,b]



[a]Reaction condition: **1** (0.3 mmol), **2** (0.2 mmol), and K_2CO_3 (0.6 mmol) in dichloromethane (DCM; 2.0 mL) at rt for 24 h. [b]Isolated yield, only one diastereoisomer was observed by crude NMR analysis.

The developed cycloaddition reaction could be performed on a gram scale. Under the optimal reaction conditions, the preparative scale reaction of **1a** and **2r** proceeded smoothly and gave the product **3ar** in 80% yield, which could be further derived by Suzuki coupling (Scheme 6). In addition, treating pyrazole **5n** with KOH, oxalyl chloride, and piperdin-1-amine, could produce hydrazide **7** in 79% yield over three steps.

A plausible mechanism was proposed, as shown in Scheme 7. Intermediate A was first generated from the corresponding hydrazonyl chloride **1a** by dechlorination. Then, the nitrile imine B formed in the presence of a base by deprotonation reacted with enamide **2a** through either a concerted or stepwise process to generate the desired product. The pyrazole **5o** could be obtained by $AlCl_3$-facilitated β-elimination. This mechanistic scheme was supported by the detection of key intermediates A by high-resolution mass spectrometry (HRMS).

CONCLUSIONS

In summary, we have developed the first [3 + 2] cycloadditions of in situ generated nitrile imines with enamides. A very wide range of nitrile imines and enamides were compatible with the mild reaction conditions and provided the highly functionalized pyrazolines. In addition, the products could be efficiently converted to useful pyrazoles via a one-pot process. This protocol expands the chemistry of both enamides and nitrile imines. The potential application of these compounds is under investigation in our laboratory.

EXPERIMENTAL SECTION

General Information. Unless otherwise specified, all reactions were carried out under a nitrogen atmosphere in anhydrous conditions. All chemicals and solvents which are commercially available were used without further purification. Analytical thin-layer chromatography (TLC) was performed on silica gel plates (GF-254) using UV light (254 and 365 nm). Flash chromatography was conducted on silica gel (200–

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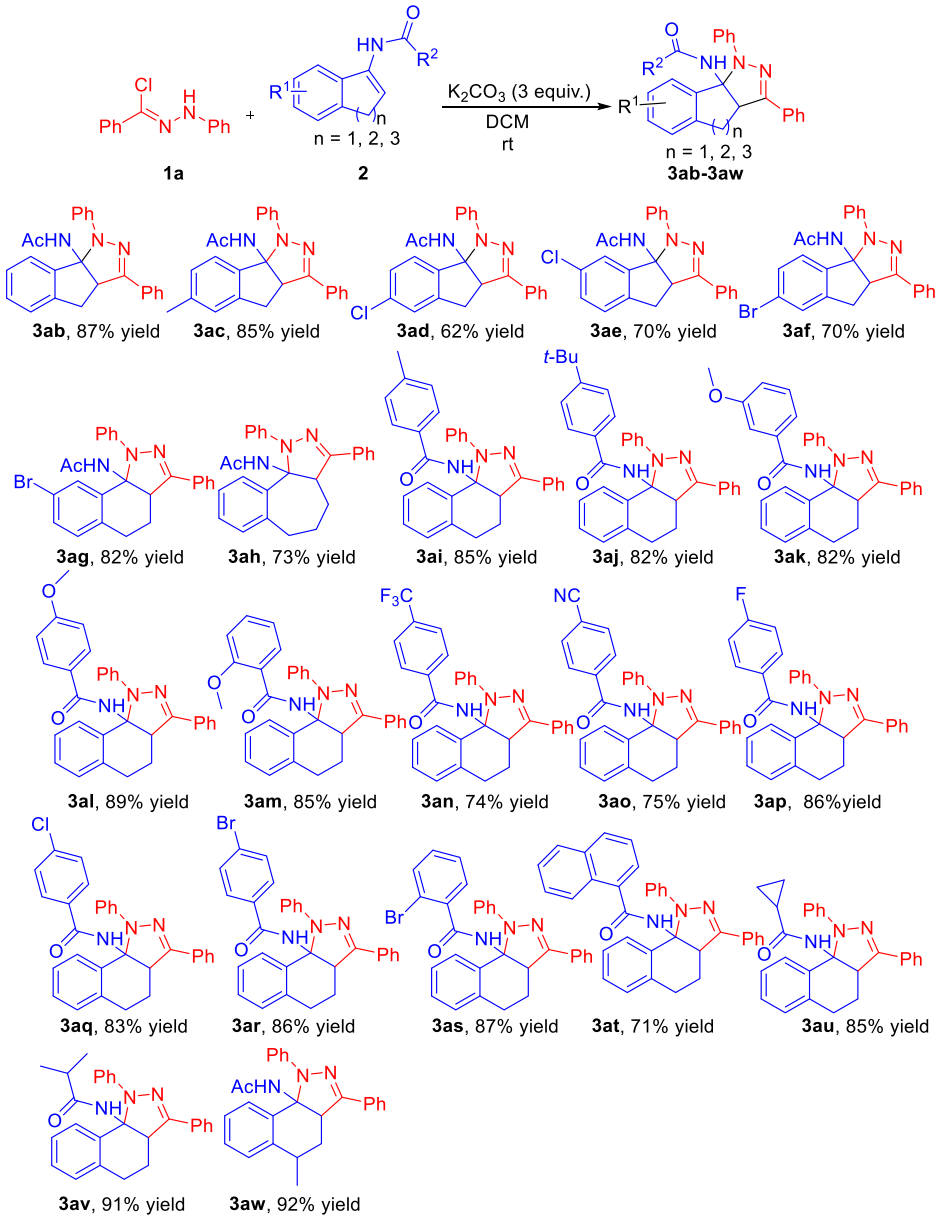
Scheme 3. Substrate Scope of Cyclic Enamides[a,b]



[a]Reaction condition: **1** (0.3 mmol), **2** (0.2 mmol), and K_2CO_3 (0.6 mmol) in DCM (2.0 mL) at rt for 24 h. [b]Isolated yield, only one diastereoisomer was observed by crude NMR analysis.

300 mesh). NMR spectra were recorded at ambient temperature in $CDCl_3$ on a Bruker Ascend III TM600 NMR spectrometer (^{1}H NMR at 600 MHz and $^{13}C\{^1H\}$ NMR at 150 MHz) and a Bruker Avance NEO NMR spectrometer (^{1}H NMR at 500 MHz and $^{13}C\{^1H\}$ NMR at 125 MHz). Chemical shifts were reported in parts per million (ppm) downfield from an internal standard, tetramethylsilane (0 ppm). All high-resolution mass spectra were obtained on an Agilent 6200 Q-TOF MS. Melting points were determined on a WRX-4 melting point apparatus. Enamides[16a] and hydrazonoyl chloride[13d,17] were prepared according to the literature procedures.

General Procedure for the Synthesis of Pyrazolines. To a dried test tube with a magnetic stirring bar under N_2 at room temperature were added hydrazonoyl chloride **1** (0.3 mmol) and enamide **2** (0.20 mmol), followed by the addition of K_2CO_3 (0.6 mmol). Then, dichloromethane (2.00 mL) was introduced by a syringe and the mixture was stirred at room temperature for 24 h. The solvent was removed by rotary evaporation, and the residue was purified by column chromatography on silica gel to afford the desired product.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)acetamide (**3aa**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (73.2 mg, yield: 96%); mp 180−181 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.82 (d, J = 7.3 Hz, 2H), 7.43 (t, J = 7.5 Hz, 2H), 7.38 (d, J = 7.3 Hz, 1H), 7.36−7.32 (m, 1H), 7.23−7.20 (m, 2H), 7.12 (t, J = 7.8 Hz, 3H), 7.00−6.89 (m, 3H), 6.32 (s, 1H), 4.45 (d, J = 7.8 Hz, 1H), 2.62−2.50 (m, 2H), 2.13 (d, J = 13.3 Hz, 1H), 1.95 (s, 3H), 1.93−1.87 (m, 1H); $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 170.1, 153.8, 141.9, 140.9, 133.2, 131.8, 129.1, 128.9, 128.7, 128.18, 128.0, 127.3, 126.6, 125.9, 123.4, 120.7, 82.8, 52.9, 27.8, 26.0, 23.9; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{25}H_{24}N_3O$, 382.1914; found, 382.1911.

*N-((3a,9b)-3-Phenyl-1-(p-tolyl)-1,3a,4,5-tetrahydro-9bH-benzo-[g]indazol-9b-yl)acetamide (**3ba**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (72.7 mg, yield: 92%); mp 177−178 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.73 (d, J = 7.2 Hz, 2H), 7.35−

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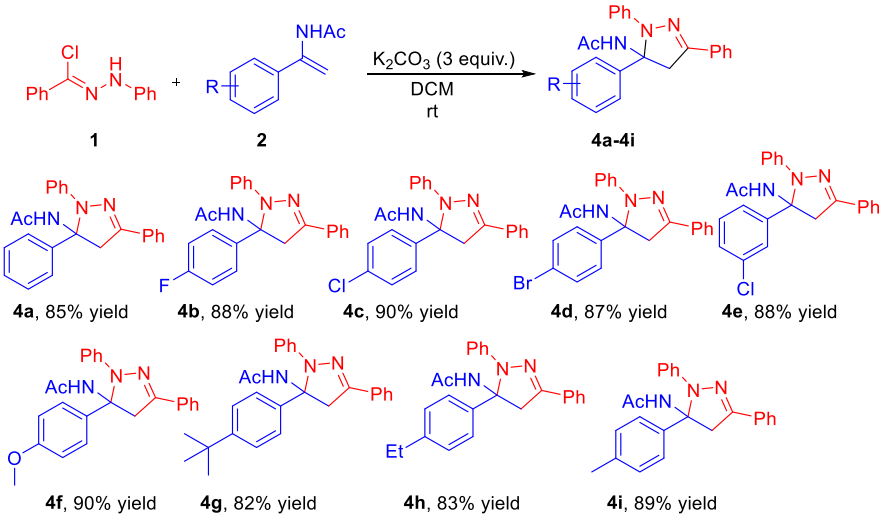
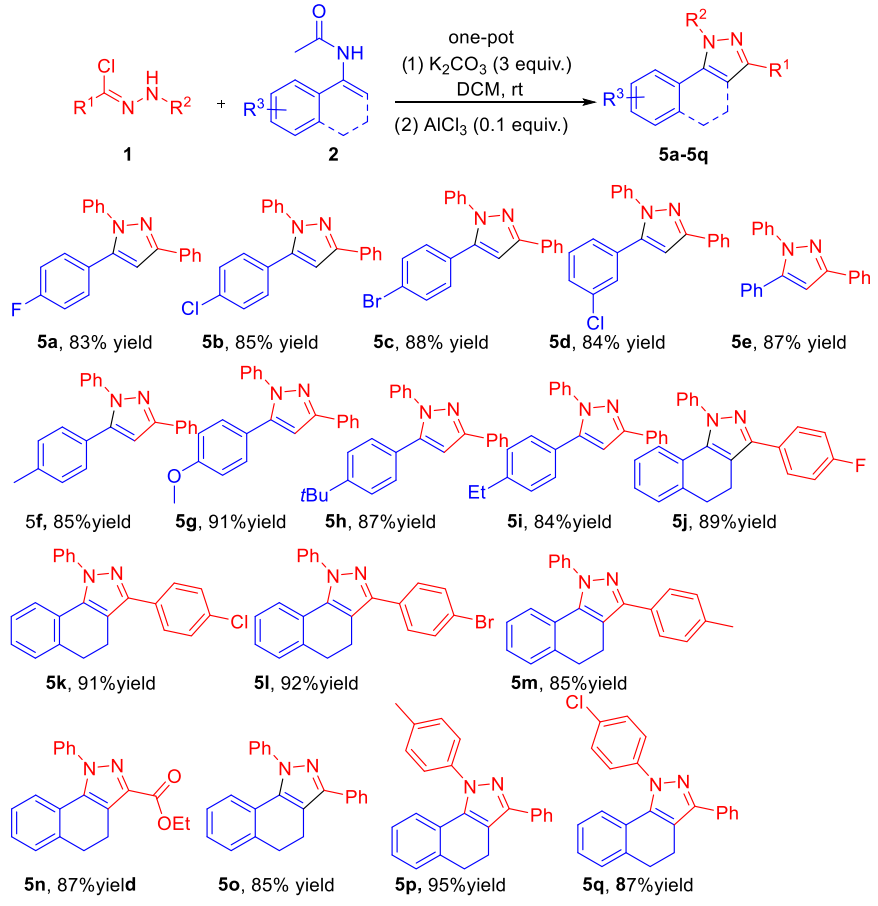
Scheme 4. Substrate Scope of Acyclic Enamides[a,b]



[a]Reaction condition: **1** (0.3 mmol), **2** (0.2 mmol), and K_2CO_3 (0.6 mmol) in DCM (2.0 mL) at rt for 24 h. [b]Isolated yield.

Scheme 5. One-Pot Preparation of Pyrazoles[a,b]



[a]Reaction condition: (1) **1** (0.3 mmol), **2** (0.2 mmol), and K_2CO_3 (0.6 mmol) in DCM (2.0 mL) at rt for 24 h; (2) $AlCl_3$ (0.02 mmol), 0.5 h. [b]Isolated yield.

7.27 (m, 3H), 7.20−7.17 (m, 1H), 7.15−7.11 (m, 2H), 7.07−7.05 (m, 1H), 6.85 (d, J = 8.2 Hz, 2H), 6.67 (d, J = 8.4 Hz, 2H), 6.15 (s, 1H), 4.36 (dd, J = 8.3, 1.5 Hz, 1H), 2.55 (dd, J = 8.2, 3.5 Hz, 2H), 2.15 (s, 3H), 2.07−2.03 (m, 1H), 1.86 (s, 3H), 1.87−1.81 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 168.9, 152.7, 139.8, 138.3, 132.5, 132.4, 130.8, 128.2, 127.9, 127.7, 127.0, 126.8, 126.0, 125.4, 124.8, 120.4, 81.7, 51.5, 26.8, 24.8, 22.8, 19.7; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{26}H_{26}N_3O$, 396.2070; found, 396.2070.

*N-((3a,9b)-3-Phenyl-1-(m-tolyl)-1,3a,4,5-tetrahydro-9bH-benzo-[g]indazol-9b-yl)acetamide (**3ca**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (71.9 mg, yield: 91%); mp 178−

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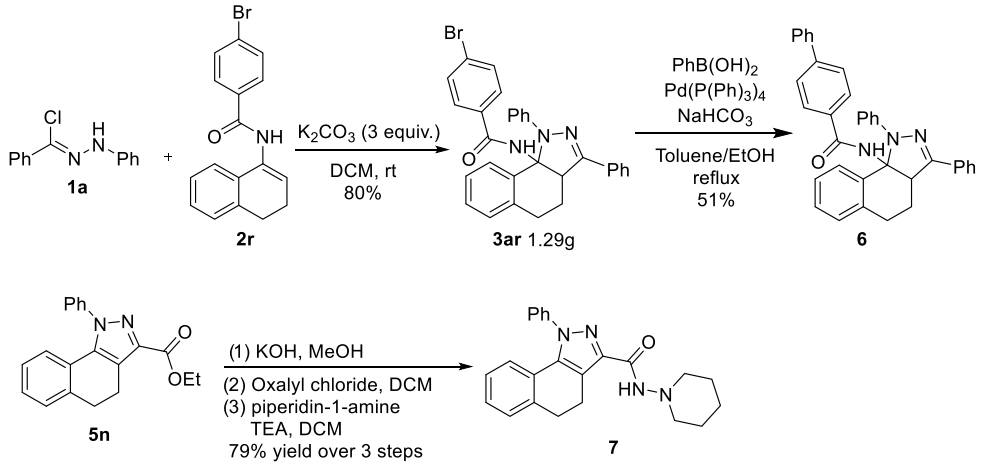
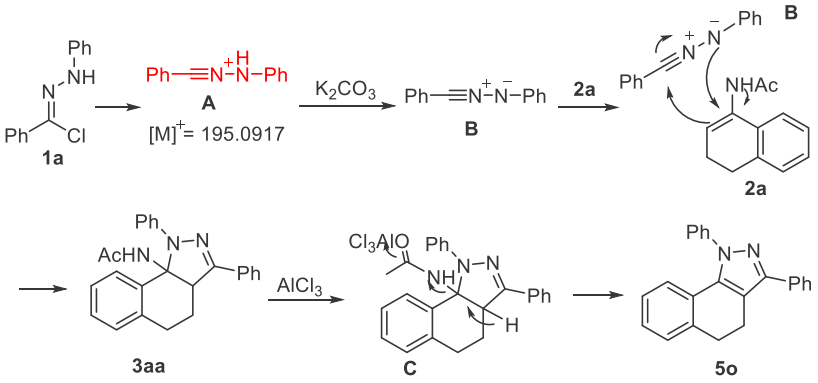
Scheme 6. Gram-Scale Synthesis and Derivation



Scheme 7. Plausible Reaction Mechanism



179 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.78−7.72 (m, 2H), 7.37−7.34 (m, 2H), 7.32−7.28 (m, 1H), 7.26−7.22 (m, 1H), 7.16−7.13 (m, 2H), 7.08−7.05 (m, 1H), 6.93 (t, J = 7.8 Hz, 1H), 6.70 (d, J = 7.5 Hz, 1H), 6.66 (s, 1H), 6.59 (d, J = 8.0 Hz, 1H), 6.14 (s, 1H), 4.38 (d, J = 8.5 Hz, 1H), 2.56−2.52 (m, 2H), 2.12 (s, 3H), 2.09−2.01 (m, 1H), 1.89 (s, 3H), 1.87−1.80 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 168.9, 152.7, 140.7, 139.9, 137.5, 132.4, 130.8, 128.0, 127.7, 127.4, 127.1, 126.9, 126.2, 125.5, 124.7, 123.3, 120.6, 116.8, 81.7, 51.5, 26.7, 24.9, 22.9, 20.4; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{26}H_{26}N_3O$, 396.2070; found, 396.2069.

*N-((3a,9b)-1-(4-Chlorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3da**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (68.1 mg, yield: 82%); mp 189−190 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.81−7.79 (m, 2H), 7.46−7.34 (m, 5H), 7.24−7.19 (m, 2H), 7.07−7.03 (m, 2H), 6.89−6.83 (m, 2H), 6.38 (s, 1H), 4.41 (d, J = 8.2 Hz, 1H), 2.60−2.46 (m, 2H), 2.19−2.08 (m, 1H), 1.96 (s, 3H), 1.91−1.85 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.2, 154.1, 140.9, 140.5, 132.7, 131.5, 129.4, 128.9, 128.6, 128.4, 128.1, 127.3, 126.6, 125.9, 121.2, 114.9, 82.7, 53.1, 27.6, 25.9, 23.8; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{25}H_{23}ClN_3O$, 416.1524; found, 416.1526.

*N-((3a,9b)-1-(3-Chlorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ea**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (68.1 mg, yield: 82%); mp 185−186 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.74 (d, J = 7.0 Hz, 2H), 7.39−7.32 (m, 3H), 7.20−7.13 (m, 3H), 7.03 (d, J = 7.2 Hz, 1H), 6.98 (t, J = 2.0 Hz, 1H), 6.90 (t, J = 8.1 Hz, 1H), 6.77 (d, J = 7.8 Hz, 1H), 6.70 (d, J = 6.8 Hz, 1H), 6.30 (s, 1H), 4.35 (d, J = 8.3 Hz, 1H), 2.49 (d, J = 16.3 Hz, 1H), 2.39 (t, J = 13.5 Hz, 1H), 2.03 (d, J = 12.4 Hz, 1H), 1.90 (s, 3H), 1.85−1.74 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 169.2, 153.0, 142.0, 139.8, 133.2, 131.3, 130.4, 128.4, 127.8, 127.3, 127.0, 126.5, 126.2, 125.6, 124.8, 121.4, 118.4, 116.0, 81.6, 52.2, 26.5, 25.0, 22.8; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{25}H_{23}ClN_3O$, 416.1524; found: 416.1523.

*N-((3a,9b)-3-Phenyl-1-(o-tolyl)-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3fa**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (68.8 mg, yield: 87%); mp 166−167 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.71 (d, J = 7.4 Hz, 2H), 7.33 (t, J = 7.4 Hz, 2H), 7.29 (d, J = 7.1 Hz, 1H), 7.16−7.08 (m, 5H), 7.03 (d, J = 7.1 Hz, 1H), 6.95 (t, J = 7.2 Hz, 1H), 6.69 (d, J = 7.8 Hz, 1H), 6.32 (s, 1H), 4.41 (t, J = 6.5 Hz, 1H), 2.84−2.76 (m, 2H), 2.11−2.06 (m, 2H), 1.84 (s, 3H), 1.40 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 169.7, 154.8, 140.0, 139.8, 137.7, 133.9, 131.9, 129.0, 128.8, 128.2 128.1, 128.0, 127.4, 126.6, 126.3, 126.2, 126.0, 82.8, 51.5, 28.4, 25.0, 24.1, 17.7; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{26}H_{26}N_3O$, 396.2070; found, 396.2070.

*N-((3a,9b)-1-(2-Chlorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ga**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (35.7 mg, yield: 43%); mp 170−171 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.79−7.78 (m, 2H), 7.44−7.37 (m, 4H), 7.28 (dd, J = 1.4 Hz, 1H), 7.15 (d, J = 8.1 Hz, 1H), 7.04−7.00 (m, 3H), 6.97−6.94 (m, 2H), 6.62 (dd, J = 7.9, 1.5 Hz, 1H), 4.41 (dd, J = 6.9, 3.3 Hz, 1H), 2.71−2.64 (m, 1H), 2.60−2.53 (m, 1H), 2.25−2.17 (m, 2H), 2.05 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 169.2, 154.2, 140.3, 139.2, 133.0, 131.8, 131.7, 130.0, 129.8, 129.1, 128.8, 128.0, 127.9, 127.8, 127.0, 126.7, 126.1, 126.0, 83.7, 53.9, 27.7, 24.5, 23.9; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{25}H_{23}ClN_3O$, 416.1524; found, 416.1523.

*N-((3a,9b)-1-(2-Chloro-4-fluorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ha**).* The product was obtained by column chromatography on silica gel (eluent:

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petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (30.3 mg, yield: 35%); mp 190−191 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.69 (d, J = 7.2 Hz, 2H), 7.36−7.30 (m, 3H), 7.10 (s, 1H), 7.04 (d, J = 7.8 Hz, 1H), 7.02−6.95 (m, 3H), 6.90 (t, J = 7.4 Hz, 1H), 6.63−6.57 (m, 1H), 6.49 (dd, J = 8.8, 5.7 Hz, 1H), 4.32 (dd, J = 6.9, 3.6 Hz, 1H), 2.61 (d, J = 14.1 Hz, 1H), 2.49−2.44 (m, 1H), 2.18−2.09 (m, 2H), 1.98 (s, 3H); ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 168.2, 159.5 (d, J = 250.5 Hz), 153.3, 138.0, 135.7 (d, J = 3.7 Hz), 131.9, 131.7 (d, J = 10.8 Hz), 130.5, 129.9 (d, J = 9.2 Hz), 128.1, 127.8, 127.1, 127.0, 125.6, 125.2, 124.8, 116.0 (d, J = 25.8 Hz), 113.2 (d, J = 21.9 Hz), 82.5, 52.9, 26.6, 23.5, 22.8; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{25}H_{21}ClFN_3NaO$, 456.1249; found, 456.1249.

*N-((3a,9b)-1-(2,4-Difluorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ia**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (50.1 mg, yield: 60%); mp 188−189 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.79 (d, J = 7.0 Hz, 2H), 7.46−7.35 (m, 3H), 7.26−7.24 (m, 1H), 7.07−6.99 (m, 2H), 6.96 (d, J = 8.1 Hz, 1H), 6.86−6.79 (m, 2H), 6.69−6.54 (m, 2H), 4.44 (d, J = 5.5 Hz, 1H), 2.58 (d, J = 15.1 Hz, 1H), 2.35 (t, J = 14.0 Hz, 1H), 2.23−2.16 (m, 1H), 2.09 (s, 3H), 2.08−2.02 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 169.4, 161.3, 161.2, 159.3, 159.2, 157.6, 157.5, 155.6, 155.5, 154.6, 139.4, 132.2, 131.5, 129.3, 128.9, 128.1, 127.9, 127.5, 127.4, 126.8, 126.1, 125.9, 125.8, 111.1, 111.1, 110.9, 110.9, 104.0, 103.8, 103.6, 82.7, 54.0, 27.5, 24.5, 23.9; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{25}H_{21}F_2N_3NaO$, 440.1545; found, 440.1544.

*N-((3a,9b)-1-(2,4-Dichlorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ja**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a yellow solid (37.7 mg, yield: 42%); mp 189−190 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.70 (d, J = 6.8 Hz, 2H), 7.39−7.30 (m, 3H), 7.23 (d, J = 2.4 Hz, 1H), 7.09 (d, J = 7.7 Hz, 1H), 7.07−7.00 (m, 2H), 6.99−6.91 (m, 2H), 6.87 (dd, J = 8.7, 2.5 Hz, 1H), 6.47 (d, J = 8.6 Hz, 1H), 4.40−4.24 (m, 1H), 2.63−2.58 (m, 1H), 2.51−2.41 (m, 1H), 2.21−2.07 (m, 2H), 1.99 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 168.2, 153.7, 138.1, 131.7, 130.5, 129.6, 129.3, 128.7, 127.8, 127.6, 127.1, 126.7, 126.4, 126.2, 125.7, 125.3, 124.9, 120.3, 82.6, 53.0, 26.7, 23.57, 22.9; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{25}H_{21}Cl_2N_3NaO$, 472.0954; found, 472.0955.

*N-((3a,9b)-1-(3,4-Dichlorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ka**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (60.2 mg, yield: 67%); mp 199−200 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.74 (d, J = 6.9 Hz, 2H), 7.40−7.34 (m, 3H), 7.33 (d, J = 6.3 Hz, 1H), 7.21−7.15 (m, 3H), 7.08 (d, J = 2.6 Hz, 1H), 7.03 (dd, J = 14.3, 7.9 Hz, 2H), 6.70−6.65 (m, 1H), 6.27 (s, 1H), 4.35 (d, J = 8.4 Hz, 1H), 2.50 (d, J = 14.6 Hz, 1H), 2.40−2.26 (m, 1H), 2.03 (d, J = 13.4 Hz, 1H), 1.93 (s, 3H), 1.86−1.76 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 169.2, 153.4, 140.3, 139.8, 131.3, 131.0, 130.2, 128.9, 128.5, 127.9, 127.5, 127.2, 126.4, 125.7, 124.9, 124.5, 119.6, 117.0, 81.6, 52.4, 26.5, 25.0, 22.8; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{25}H_{22}Cl_2N_3O$, 450.1134; found, 450.1133.

*N-((3a,9b)-1-(3,5-Dichlorophenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3la**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (71.0 mg, yield: 79%); mp 210−211 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.84 (d, J = 8.0 Hz, 2H), 7.52 (d, J = 7.6 Hz, 1H), 7.49−7.41 (m, 3H), 7.33 (t, J = 7.3 Hz, 1H), 7.28 (d, J = 1.3 Hz, 1H), 7.15 (d, J = 8.9 Hz, 1H), 6.93 (d, J = 1.8 Hz, 2H), 6.85 (t, J = 1.8 Hz, 1H), 6.07 (s, 1H), 4.48 (d, J = 8.5 Hz, 1H), 2.59 (d, J = 14.8 Hz, 1H), 2.50−2.38 (m, 1H), 2.16−2.11 (m, 1H), 2.10 (s, 3H), 1.96−1.85 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.0, 154.6, 143.62, 140.9, 134.8, 131.7, 131.1, 129.7, 128.9, 128.7, 128.2, 127.5, 126.8, 126.1, 121.7, 116.4, 82.7, 53.4, 27.5, 26.2, 24.0; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{25}H_{21}Cl_2N_3NaO$, 472.0954; found, 472.0956.

*N-((3a,9b)-1-(3,5-Dimethylphenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ma**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (75.3 mg, yield: 92%); mp 189−190 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.75 (d, J = 7.3 Hz, 2H), 7.35 (t, J = 7.5 Hz, 2H), 7.30 (d, J = 7.3 Hz, 1H), 7.23 (d, J = 8.5 Hz, 1H), 7.18−7.12 (m, 2H), 7.08 (d, J = 7.2 Hz, 1H), 6.55 (s, 1H), 6.41 (s, 2H), 6.12 (s, 1H), 4.38 (d, J = 8.3 Hz, 1H), 2.56−2.54 (m, 2H), 2.08 (s, 6H), 2.06−2.01 (m, 1H), 1.89 (s, 3H), 1.88−1.83 (m, 1H); ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 168.9, 152.6, 140.6, 139.9, 137.2, 132.5, 130.8, 127.9, 127.7, 127.1, 126.9, 126.3, 125.5, 124.5, 124.4, 117.8, 81.7, 51.3, 28.9, 26.7, 24.9, 22.9, 20.3; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{27}H_{27}N_3NaO$, 432.2046; found, 432.2046.

*N-((3a,9b)-1-(3-Chloro-4-methylphenyl)-3-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3na**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a yellow solid (50.6 mg, yield: 59%); mp 201−202 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.74 (d, J = 7.4 Hz, 2H), 7.36 (t, J = 7.4 Hz, 2H), 7.30 (dd, J = 14.8, 7.5 Hz, 2H), 7.18−7.14 (m, 2H), 7.05 (d, J = 6.2 Hz, 1H), 6.93 (s, 1H), 6.83 (d, J = 8.3 Hz, 1H), 6.55 (d, J = 8.2 Hz, 1H), 6.19 (s, 1H), 4.35 (d, J = 8.2 Hz, 1H), 2.53−2.42 (m, 2H), 2.15 (s, 3H), 2.03 (d, J = 13.1 Hz, 1H), 1.90 (s, 3H), 1.86−1.81 (m, 1H); ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 169.0, 153.0, 139.8, 139.8, 133.2, 131.8, 130.5, 129.5, 129.4, 128.2, 127.8, 127.3, 127.0, 126.2, 125.6, 124.8, 119.7, 117.3, 81.6, 52.0, 26.6, 24.9, 22.8, 18.3; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{26}H_{25}ClN_3O$, 430.1681; found, 430.1681.

*N-((3a,9b)-3-(2-Bromophenyl)-1-(3-chlorophenyl)-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3oa**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (63.1 mg, yield: 64%); mp 190−191 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.60 (d, J = 7.9 Hz, 1H), 7.47 (d, J = 7.3 Hz, 1H), 7.39 (d, J = 7.4 Hz, 1H), 7.32 (t, J = 7.4 Hz, 1H), 7.22−7.10 (m, 3H), 7.04 (d, J = 7.0 Hz, 1H), 6.99 (s, 1H), 6.93 (t, J = 8.0 Hz, 1H), 6.77 (dd, J = 18.1, 7.8 Hz, 2H), 6.29 (s, 1H), 4.63 (d, J = 7.8 Hz, 1H), 2.54−2.41 (m, 2H), 1.97 (s, 3H), 1.83−1.60 (m, 2H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 168.9, 153.9, 142.0, 139.6, 133.2, 132.9, 131.6, 131.3, 130.3, 129.5, 128.4, 127.3, 127.2, 126.5, 126.5, 124.9, 121.7, 121.4, 118.5, 116.2, 81.3, 54.6, 26.5, 24.3, 22.8; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{25}H_{22}BrClN_3O$, 494.0629; found, 494.0630.

*N-((3a,9b)-1-Phenyl-3-(p-tolyl)-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3pa**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (68.0 mg, yield: 86%); mp 175−176 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.72 (d, J = 8.1 Hz, 3H), 7.35−7.33 (m, 1H), 7.24−7.21 (m, 3H), 7.16−7.09 (m, 3H), 6.96 (t, J = 7.2 Hz, 1H), 6.91 (d, J = 3.4 Hz, 2H), 6.21 (s, 1H), 4.45 (d, J = 9.0 Hz, 1H), 2.64−2.59 (m, 2H), 2.40 (s, 3H), 2.13 (d, J = 13.3 Hz, 1H), 1.98 (s, 3H), 1.95−1.87 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.0, 154.0, 142.0, 140.9, 133.3, 129.5, 129.4, 129.2, 128.9, 128.7, 128.1, 128.0, 127.2, 126.5, 125.8, 123.3, 120.6, 113.6, 82.7, 52.8, 27.7, 26.0, 23.9, 21.4; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{26}H_{26}N_3O$, 396.2070; found, 396.2070.

*N-((3a,9b)-3-(4-Fluorophenyl)-1-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3qa**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (65.5 mg, yield: 82%); mp 217−218 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.79 (dd, J = 8.8, 5.4 Hz, 2H), 7.356−7.31 (m, 1H), 7.22−7.20 (m, 2H), 7.14−7.09 (m, 5H), 6.95 (t, J = 7.3 Hz, 1H), 6.90 (d, J = 7.7 Hz, 2H), 6.34 (s, 1H), 4.41 (d, J = 7.7 Hz, 1H), 2.66−2.49 (m, 2H), 2.08 (d, J = 13.5 Hz, 1H), 1.94 (s, 3H), 1.93−1.85 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.1, 163.3 (d, J = 249.7 Hz), 152.9, 141.9, 140.8, 133.1, 128.7, 128.4 (d, J = 8.2 Hz), 128.2, 128.1 (d, J = 3.3 Hz), 128.0, 127.3, 125.9, 123.5, 120.7, 115.9 (d, J = 21.8 Hz), 82.8, 52.9, 27.7, 25.9, 23.8; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{25}H_{23}FN_3O$, 400.1820; found, 400.1819.

*N-((3a,9b)-3-(4-Chlorophenyl)-1-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ra**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (66.4 mg, yield: 80%); mp 167−168 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.74 (d, J = 8.6 Hz, 2H), 7.39 (d, J = 8.6 Hz, 2H), 7.35−7.31 (m, 1H), 7.23−7.21 (m, 2H),

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7.17−7.10 (m, 3H), 6.97 (t, J = 7.3 Hz, 1H), 6.90 (d, J = 7.7 Hz, 2H), 6.26 (s, 1H), 4.41 (d, J = 7.8 Hz, 1H), 2.67−2.52 (m, 2H), 2.07 (d, J = 13.4 Hz, 1H), 1.96 (s, 3H), 1.95−1.86 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 170.1, 152.7, 141.7, 140.8, 134.9, 133.1, 130.4, 129.0, 128.8, 128.3, 128.0, 127.7, 127.3, 125.9, 123.6, 120.7, 82.9, 52.7, 27.7, 25.9, 23.9; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{25}H_{23}ClN_3O$, 416.1524; found, 416.1524.

*N-((3a,9b)-3-(4-Bromophenyl)-1-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3sa**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (79.9 mg, yield: 87%); mp 170−171 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.68 (d, J = 8.5 Hz, 2H), 7.56 (d, J = 8.5 Hz, 2H), 7.35−7.31 (m, 1H), 7.25−7.22 (m, 2H), 7.16−7.13 (m, 3H), 6.98 (t, J = 7.3 Hz, 1H), 6.90 (d, J = 7.8 Hz, 2H), 6.19 (s, 1H), 4.42 (d, J = 7.9 Hz, 2H), 2.70−2.51 (m, 2H), 2.08 (d, J = 12.1 Hz, 1H), 1.99 (s, 3H), 1.95−1.86 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 170.0, 152.8, 141.7, 140.8, 133.1, 132.0, 130.8, 129.5, 128.8, 128.3, 128.0, 127.9, 127.2, 126.0, 123.7, 123.2, 120.8, 82.9, 52.6, 27.7, 25.9, 23.9; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{25}H_{23}BrN_3O$, 460.1019; found, 460.1019.

*Ethyl (3a,9b)-9b-Acetamido-1-phenyl-3a,4,5,9b-tetrahydro-1H-benzo[g]indazole-3-carboxylate (**3ta**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (65.6 mg, yield: 87%); mp 185−186 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.35−7.31 (m, 1H), 7.20−7.17 (m, 2H), 7.14−7.08 (m, 3H), 7.00 (t, J = 7.4 Hz, 1H), 6.91−6.87 (m, 2H), 6.58 (s, 1H), 4.47−4.26 (m, 2H), 4.14 (d, J = 7.6 Hz, 1H), 2.63 (d, J = 14.8 Hz, 1H), 2.44−2.38 (m, 1H), 2.21 (d, J = 15.1 Hz, 1H), 2.04 (s, 3H), 1.89−1.81 (m, 1H), 1.38 (t, J = 7.1 Hz, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 169.7, 162.3, 144.4, 140.7, 134.0, 131.6, 128.7, 128.5, 128.1, 127.4, 125.9, 124.9, 121.6, 83.5, 61.3, 52.7, 27.5, 25.6, 23.8, 14.4; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{22}H_{23}N_3NaO_3$, 400.1632; found, 400.1632.

*N-((3a,9b)-3-(Furan-2-yl)-1-phenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ua**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (65.3 mg, yield: 88%); mp 191−192 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.35−7.31 (m, 2H), 7.16−7.12 (m, 3H), 7.07−7.05 (m, 1H), 7.03−7.01 (m, 1H), 6.98 (t, J = 7.8 Hz, 1H), 6.86−6.81 (m, 4H), 4.23 (d, J = 8.0 Hz, 1H), 2.57−2.52 (m, 1H), 2.49−2.43 (m, 1H), 2.22−2.18 (m, 1H), 1.93 (s, 3H), 1.91−1.87 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 170.3, 150.0, 141.6, 140.5, 136.0, 132.7, 128.4, 128.1, 127.9, 127.8, 127.5, 127.0, 126.9, 125.8, 123.3, 120.8, 82.6, 54.7, 27.7, 26.1, 23.8. HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{22}N_3O_2$, 372.1707; found, 372.1705.

*N-((3a,9b)-1-Phenyl-3-(thiophen-2-yl)-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3va**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (68.5 mg, yield: 89%); mp 200−201 °C. ^1H NMR (500 MHz, $CDCl_3$) δ: 7.55 (d, J = 1.9 Hz, 1H), 7.36−7.29 (m, 1H), 7.24−7.18 (m, 2H), 7.17−7.05 (m, 3H), 6.96 (t, J = 7.3 Hz, 1H), 6.91−6.84 (m, 2H), 6.69 (d, J = 3.5 Hz, 1H), 6.53 (dd, J = 3.5, 1.8 Hz, 1H), 6.21 (s, 1H), 4.47−4.23 (m, 1H), 2.76−2.55 (m, 2H), 2.23−2.19 (m, 1H), 2.01 (s, 3H), 1.99−1.86 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 169.9, 147.8, 146.4, 143.7, 141.6, 140.7, 132.9, 128.6, 128.2, 127.9, 127.2, 125.9, 123.7, 121.0, 111.7, 110.4, 82.2, 53.4, 27.7, 26.4, 23.9. HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{22}N_3OS$, 388.1478; found, 388.1478.

*3-Methyl-1-phenyl-4,5-dihydro-1H-benzo[g]indazole (**3wa**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1) as a light yellow solid (40.1 mg, yield: 77%); mp 113−114 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.53−7.42 (m, 4H), 7.42−7.36 (m, 1H), 7.27 (dd, J = 7.6, 1.3 Hz, 1H), 7.12 (td, J = 7.5, 1.3 Hz, 1H), 6.98 (td, J = 7.7, 1.3 Hz, 1H), 6.83 (dd, J = 7.9, 1.3 Hz, 1H), 2.98 (dd, J = 8.4, 6.4 Hz, 2H), 2.77−2.57 (m, 2H), 2.31 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 146.0, 140.9, 138.0, 137.2, 129.2, 128.6, 127.8, 127.3, 127.2, 126.3, 125.6, 123.1, 118.6, 30.7, 19.3, 11.8. HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{18}H_{17}N_2$, 261.1386; found, 261.1385.

*N-((3a,8b)-1,3-Diphenyl-3a,4-dihydroindeno[1,2-c]pyrazol-8b-(1H)-yl)acetamide (**3ab**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a yellow solid (63.9 mg, yield: 87%); mp 236−237 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.74 (d, J = 7.2 Hz, 2H), 7.42−7.28 (m, 3H), 7.29 (d, J = 7.4 Hz, 2H), 7.24−7.19 (m, 2H), 7.10 (t, J = 7.1 Hz, 1H), 7.03 (t, J = 7.3 Hz, 1H), 6.49 (s, 1H), 4.77 (dd, J = 9.8, 4.5 Hz, 1H), 3.64 (dd, J = 16.5, 9.8 Hz, 1H), 3.20 (dd, J = 16.6, 4.4 Hz, 1H), 1.96 (s, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 169.2, 152.0, 143.3, 142.6, 140.1, 132.1, 129.3, 128.8, 128.6, 126.9, 126.1, 125.2, 123.8, 123.0, 120.0, 92.4, 56.6, 36.6, 23.8; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{24}H_{22}N_3O$, 368.1757; found, 368.1756.

*N-((3a,8b)-6-Methyl-1,3-diphenyl-3a,4-dihydroindeno[1,2-c]-pyrazol-8b(1H)-yl)acetamide (**3ac**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (64.8 mg, yield: 85%); mp 160−161 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.73 (d, J = 7.4 Hz, 2H), 7.41−7.33 (m, 5H), 7.29−7.27 (m, 2H), 7.10 (d, J = 8.0 Hz, 1H), 7.04−6.98 (m, 2H), 6.89 (d, J = 7.9 Hz, 1H), 6.55 (s, 1H), 4.73 (dd, J = 9.6, 4.1 Hz, 1H), 3.64−3.52 (m, 1H), 3.14 (dd, J = 16.5, 4.1 Hz, 1H), 2.26 (s, 3H), 1.95 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 169.2, 151.9, 143.3, 142.8, 139.3, 137.3, 132.1, 128.8, 128.6, 128.5, 127.8, 126.1, 125.7, 123.5, 122.7, 119.7, 92.1, 56.7, 36.4, 23.8, 21.3; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{25}H_{23}N_3NaO$, 404.1733; found, 404.1732.

*N-((3a,8b)-6-Chloro-1,3-diphenyl-3a,4-dihydroindeno[1,2-c]-pyrazol-8b(1H)-yl)acetamide (**3ad**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (49.7 mg, yield: 62%); mp 174−175 °C. ^1H NMR (600 MHz, $CDCl_3$): δ 7.70 (d, J = 7.3 Hz, 2H), 7.41 (t, J = 7.4 Hz, 2H), 7.37 (d, J = 7.3 Hz, 1H), 7.29 (d, J = 7.7 Hz, 2H), 7.25 (t, J = 7.8 Hz, 2H), 7.13 (s, 1H), 7.07−6.96 (m, 3H), 6.76 (s, 1H), 4.63 (dd, J = 9.8, 4.2 Hz, 1H), 3.61 (dd, J = 16.7, 9.9 Hz, 1H), 3.10 (dd, J = 16.7, 4.0 Hz, 1H), 1.95 (s, 3H); $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 169.5, 152.4, 144.6, 142.9, 138.3, 135.0, 131.7, 128.9, 128.7, 126.9, 126.1, 125.2, 124.9, 123.4, 120.2, 91.8, 57.3, 36.3, 23.7; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{24}H_{20}ClN_3NaO$, 424.1187; found, 424.1187.

*N-((3a,8b)-7-Chloro-1,3-diphenyl-3a,4-dihydroindeno[1,2-c]-pyrazol-8b(1H)-yl)acetamide (**3ae**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (56.2 mg, yield: 70%); mp 180−181 °C. ^1H NMR (600 MHz, $CDCl_3$): δ 7.71 (d, J = 8.4 Hz, 2H), 7.41 (t, J = 7.4 Hz, 2H), 7.37 (d, J = 7.3 Hz, 1H), 7.32−7.27 (m, 4H), 7.16 (dd, J = 8.1, 1.7 Hz, 1H), 7.12−7.03 (m, 3H), 6.71 (s, 1H), 4.68 (dd, J = 9.8, 4.3 Hz, 1H), 3.60 (dd, J = 16.6, 9.9 Hz, 1H), 3.10 (dd, J = 16.6, 4.2 Hz, 1H), 1.96 (s, 3H); $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$) δ 169.5, 152.5, 142.8, 141.6, 141.0, 132.3, 131.7, 129.3, 129.0, 128.9, 128.7, 126.1, 126.1, 124.1, 123.6, 120.2, 92.1, 57.4, 36.2, 23.7; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{24}H_{21}ClN_3O$, 402.1368; found, 402.1368.

*N-((3a,8b)-6-Bromo-1,3-diphenyl-3a,4-dihydroindeno[1,2-c]-pyrazol-8b(1H)-yl)acetamide (**3af**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (62.3 mg, yield: 70%); mp 187−188 °C. ^1H NMR (500 MHz, DMSO-d_6): δ 9.03 (s, 1H), 7.65 (d, J = 7.4 Hz, 2H), 7.37 (dd, J = 14.8, 7.6 Hz, 6H), 7.33−7.28 (m, 2H), 7.20 (t, J = 7.9 Hz, 2H), 6.83 (t, J = 7.2 Hz, 1H), 4.66 (dd, J = 9.8, 3.4 Hz, 1H), 3.56 (dd, J = 17.4, 9.9 Hz, 1H), 2.95 (d, J = 17.5 Hz, 1H), 1.84 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, DMSO-d_6): δ 169.7, 145.3, 143.0, 139.8, 132.3, 130.0, 129.2, 128.8, 128.7, 128.5, 127.1, 126.2, 123.0, 121.1, 117.0, 90.6, 55.6, 36.0, 23.5; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{24}H_{20}BrN_3NaO$, 468.0682; found, 468.0684.

*N-((3a,9b)-8-Bromo-1,3-diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3ag**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a yellow solid (75.3 mg, yield: 82%); mp 185−186 °C. ^1H NMR (500 MHz, DMSO-d_6): δ 9.01 (s, 1H), 7.74−7.72 (m, 2H), 7.53 (d, J = 2.0 Hz, 1H), 7.39 (t, J = 7.5 Hz, 2H), 7.35−7.31 (m, 1H), 7.20 (dd, J = 7.9, 2.0 Hz, 1H), 7.05−7.01 (m, 2H), 6.95 (d, J = 7.5 Hz, 2H), 6.91 (d, J = 8.0 Hz, 1H), 6.74 (t, J = 7.2 Hz, 1H), 4.17 (d, J = 7.8 Hz, 1H), 2.47 (s, 1H), 2.03−1.95 (m, 1H), 1.94 (s, 3H), 1.93−

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1.87 (m, 1H), 1.68−1.52 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, DMSO-d_6): δ 170.4, 150.9, 142.0, 139.8, 135.5, 132.0, 131.4, 130.5, 129.9, 129.4, 129.2, 128.4, 126.7, 122.2, 119.4, 118.7, 81.3, 54.3, 26.5, 25.8, 23.5; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{25}H_{22}BrN_3NaO$, 482.0838; found, 482.0839.

*N-((3a,10b)-1,3-Diphenyl-3a,4,5,6-tetrahydrobenzo[6,7]-cyclohepta[1,2-c]pyrazol-10b(1H)-yl)acetamide (**3ah**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a yellow solid (57.7 mg, yield: 73%); mp 201−202 °C. ^1H NMR (500 MHz, DMSO-d_6): δ 8.86 (s, 1H), 7.74−7.42 (m, 2H), 7.53 (dd, J = 7.6, 1.3 Hz, 1H), 7.39 (t, J = 7.5 Hz, 2H), 7.34−7.29 (m, 1H), 7.16−7.11 (m, 2H), 7.03 (d, J = 7.5 Hz, 1H), 6.96 (d, J = 4.3 Hz, 4H), 6.70−6.64 (m, 1H), 4.18 (d, J = 8.2 Hz, 1H), 2.27−2.14 (m, 1H), 1.92 (s, 3H), 1.81−1.74 (m, 1H), 1.45−1.38 (m, 1H), 1.08 (d, J = 6.7 Hz, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, DMSO-d_6): δ 170.3, 151.2,144.1, 142.0, 132.4, 132.1, 129.4, 129.2, 129.1, 128.3, 127.9, 126.5, 125.3, 123.9, 121.3, 118.3, 81.7, 53.3, 35.0, 29.2, 23.6, 18.5; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{26}H_{25}N_3NaO$, 418.1890; found, 418.1890.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)-4-methylbenzamide (**3ai**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (77.7 mg, yield: 85%); mp 155−156 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.83 (d, J = 7.2 Hz, 2H), 7.56 (d, J = 8.2 Hz, 2H), 7.43−7.30 (m, 4H), 7.25−7.13 (m, 7H), 7.02−6.99 (m, 3H), 6.88 (s, 1H), 4.64 (d, J = 6.9 Hz, 1H), 2.71−2.66 (m, 2H), 2.33 (s, 3H), 2.21 (d, J = 13.3 Hz, 1H), 2.14−1.97 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 166.7, 154.4, 142.7, 142.2, 141.0, 133.6, 131.8, 131.1, 129.5, 129.2, 128.9, 128.9, 128.3, 128.0, 127.3, 127.0, 126.7, 126.0, 124.2, 121.6, 83.2, 53.0, 27.9, 26.1, 21.6; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{31}H_{27}N_3NaO$, 480.2046; found, 480.2047.

*4-(tert-Butyl)-N-((3a,9b)-1,3-diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)benzamide (**3aj**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (81.9 mg, yield: 82%); mp 151−152 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.83 (d, J = 8.5 Hz, 2H), 7.61 (d, J = 8.5 Hz, 2H), 7.41−7.38 (m, 4H), 7.36−7.30 (m, 2H), 7.25−7.16 (m, 5H), 7.06−6.97 (m, 3H), 6.90 (s, 1H), 4.65 (d, J = 6.6 Hz, 1H), 2.75−2.66 (m, 2H), 2.21 (d, J = 15.0 Hz, 1H), 2.12−1.99 (m, 1H), 1.28 (s, 9H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 166.4, 155.6, 154.3, 141.9, 140.8, 133.5, 131.7, 130.9, 129.0, 128.8, 128.7, 128.1, 127.9, 127.3, 127.2, 126.7, 126.5, 125.9, 125.6, 124.1, 121.5, 83.1, 52.7, 34.9, 31.1, 27.8, 26.0; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{34}H_{33}N_3NaO$, 522.2516; found, 522.2517.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)-3-methoxybenzamide (**3ak**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (77.6 mg, yield: 82%); mp 170−171 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.84 (d, J = 7.2 Hz, 2H), 7.42 (t, J = 7.4 Hz, 2H), 7.37 (d, J = 7.2 Hz, 1H), 7.33−7.25 (m, 3H), 7.24−7.19 (m, 5H), 7.14 (d, J = 7.9 Hz, 1H), 7.06−6.97 (m, 4H), 6.91 (s, 1H), 4.63 (d, J = 9.8 Hz, 1H), 3.78 (s, 3H), 2.74−2.67 (m, 2H), 2.26−2.19 (m, 1H), 2.10−1.98 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 166.4, 159.9, 154.4, 141.9, 140.9, 135.2, 133.4, 131.7, 129.7, 129.1, 128.9, 128.8, 128.2, 128.0, 127.4, 127.1, 126.6, 126.0, 124.2, 121.6, 118.5, 111.9, 83.1, 55.4, 52.8, 27.8, 26.0; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{31}H_{27}N_3NaO_2$, 496.1995; found, 496.1997.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)-4-methoxybenzamide (**3al**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (84.2 mg, yield: 89%); mp 161−162 °C. ^1H NMR (600 MHz, $CDCl_3$): δ 7.84 (d, J = 7.4 Hz, 2H), 7.62 (d, J = 8.8 Hz, 2H), 7.42 (t, J = 7.5 Hz, 2H), 7.38 (d, J = 7.3 Hz, 1H), 7.32 (d, J = 7.4 Hz, 1H), 7.24−7.17 (m, 5H), 7.05−6.99 (m, 3H), 6.88 (d, J = 8.8 Hz, 2H), 6.79 (s, 1H), 4.63 (d, J = 7.2 Hz, 1H), 3.81 (s, 3H), 2.75−2.66 (m, 2H), 2.22 (d, J = 13.3 Hz, 1H), 2.14−1.93 (m, 1H); $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 166.2, 162.6, 154.3, 142.0, 140.9, 133.6, 131.7, 129.1, 128.9, 128.7,128.7, 128.1, 127.9, 127.2, 126.6, 126.1, 125.9, 124.1, 121.4, 113.9, 83.1, 52.9, 27.8, 26.0; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{31}H_{27}N_3NaO_2$, 496.1995; found, 496.2004.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)-2-methoxybenzamide (**3am**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (80.4 mg, yield: 85%); mp 152−153 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 8.72 (s, 1H), 8.12 (dd, J = 7.9, 1.8 Hz, 1H), 7.84 (d, J = 7.2 Hz, 2H), 7.44−7.37 (m, 4H), 7.35 (d, J = 7.3 Hz, 1H), 7.28−7.16 (m, 5H), 7.04−7.00 (m, 4H), 6.85 (d, J = 8.3 Hz, 1H), 4.71 (d, J = 7.9 Hz, 1H), 3.52 (s, 3H), 2.83−2.64 (m, 2H), 2.22 (d, J = 13.3 Hz, 1H), 2.15−1.98 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 164.8, 157.7, 154.0, 143.1, 141.3, 134.6, 133.3, 132.2, 132.0, 129.0, 128.8, 128.1, 128.0, 127.4,127.4 126.6, 126.1, 123.5, 121.3, 121.3, 121.0, 111.3, 83.6, 55.6, 52.9, 28.0, 26.2; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{31}H_{28}N_3O_2$, 474.2176; found, 474.2175.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)-4-(trifluoromethyl)benzamide (**3an**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (75.7 mg, yield: 74%); mp 166−167 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.84 (d, J = 7.2 Hz, 2H), 7.76 (d, J = 8.2 Hz, 2H), 7.66 (d, J = 8.3 Hz, 2H), 7.45−7.37 (m, 3H), 7.30−7.17 (m, 6H), 7.06 (t, J = 7.4 Hz, 1H), 6.98 (d, J = 7.6 Hz, 2H), 6.92 (s, 1H), 4.62 (d, J = 8.2 Hz, 1H), 2.73 (dd, J = 8.2, 3.7 Hz, 2H), 2.24 (d, J = 13.3 Hz, 1H), 2.12−2.02 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 165.3, 154.5, 141.8, 140.9, 137.0, 133.0, 131.6, 129.3, 129.0, 128.8, 128.4, 128.14, 127.4, 127.0, 126.6, 126.0, 125.8, 125.8, 124.4, 121.7, 83.3, 52.7, 27.8, 25.9; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{31}H_{25}F_3N_3O$, 512.1944; found, 512.1947.

*4-Cyano-N-((3a,9b)-1,3-diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)benzamide (**3ao**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (70.2 mg, yield: 75%); mp 190−191 °C. ^1H NMR (600 MHz, $CDCl_3$): δ 7.83 (d, J = 7.1 Hz, 2H), 7.74 (d, J = 8.5 Hz, 2H), 7.67 (d, J = 8.5 Hz, 2H), 7.45−7.40 (m, 2H), 7.41−7.35 (m, 1H), 7.27−7.24 (m, 2H), 7.22−7.18 (m, 4H), 7.04 (t, J = 7.4 Hz, 1H), 6.97 (d, J = 7.5 Hz, 3H), 4.57 (d, J = 10.0 Hz, 1H), 2.75−2.61 (m, 2H), 2.23 (d, J = 13.4 Hz, 1H), 2.11−1.99 (m, 1H); $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 164.8, 154.3, 141.7, 140.9, 137.5, 132.7, 132.6, 131.5, 129.29, 128.9, 128.8, 128.4, 128.1, 127.6, 126.9, 126.6, 126.0, 124.3, 121.5, 117.9, 115.5, 83.3, 52.8, 27.8, 25.9; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{31}H_{24}N_4NaO$, 491.1842; found, 491.1847.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)-4-fluorobenzamide (**3ap**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (79.3 mg, yield: 86%); mp 184−185 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.83 (d, J = 7.2 Hz, 2H), 7.66 (dd, J = 8.7, 5.3 Hz, 2H), 7.43−7.34 (m, 3H), 7.30 (d, J = 7.7 Hz, 1H), 7.27−7.14 (m, 5H), 7.08−6.95 (m, 5H), 6.87 (s, 1H), 4.60 (d, J = 8.2 Hz, 1H), 2.69 (dd, J = 8.4, 3.8 Hz, 2H), 2.21 (d, J = 11.7 Hz, 1H), 2.09−2.00 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 165.5, 165.0 (d, J = 251.3 Hz), 154.4, 141.9, 140.9, 133.3, 131.6, 130.0 (d, J = 3.2 Hz), 129.2 (d, J = 8.7 Hz), 129.2, 128.9, 128.8, 128.3, 128.0, 127.1, 126.6, 126.0, 124.2, 121.5, 115.8 (d, J = 22.0 Hz), 83.2, 52.8, 27.8, 26.0; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{30}H_{24}FN_3NaO$, 484.1796; found, 484.1797.

*4-Chloro-N-((3a,9b)-1,3-diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)benzamide (**3aq**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (79.2 mg, yield: 83%); mp 155−156 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.82 (d, J = 7.3 Hz, 2H), 7.58 (d, J = 8.4 Hz, 2H), 7.45−7.32 (m, 5H), 7.31−7.13 (m, 6H), 7.07−6.95 (m, 3H), 6.88 (s, 1H), 4.59 (d, J = 7.0 Hz, 1H), 2.71−2.68 (m, 2H), 2.21 (d, J = 14.8 Hz, 1H), 2.11−1.96 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 165.6, 154.4, 141.9, 141.0, 138.4, 133.2, 132.3, 131.7, 129.3, 129.1, 129.0, 128.9, 128.4, 128.4, 128.1, 127.2, 126.7, 126.0, 124.3, 121.6, 83.3, 52.9, 27.9, 26.0; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{30}H_{24}ClN_3NaO$, 500.1500; found, 500.1499.

*4-Bromo-N-((3a,9b)-1,3-diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)benzamide (**3ar**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl

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acetate = 10:1 to 5:1) as a white solid (89.6 mg, yield: 86%); mp 170−171 °C. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.83 (d, J = 7.1 Hz, 2H), 7.56−7.47 (m, 4H), 7.43−7.36 (m, 3H), 7.30−7.16 (m, 6H), 7.04 (t, J = 6.9 Hz, 1H), 6.97 (d, J = 7.6 Hz, 2H), 6.85 (s, 1H), 4.60 (dd, J = 8.3, 1.7 Hz, 1H), 2.70 (dd, J = 8.3, 3.6 Hz, 2H), 2.38−2.19 (m, 1H), 2.09−1.97 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 165.6, 154.3, 141.8, 140.9, 133.1, 132.6, 132.0, 131.6, 129.2, 128.9, 128.8, 128.4, 128.2, 128.0, 127.0, 126.8, 126.6, 125.9, 124.2, 121.5, 83.2, 52.8, 27.8, 25.9; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{30}H_{24}BrN_3NaO$, 544.0995; found, 544.0996.

*2-Bromo-N-((3a,9b)-1,3-diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)benzamide (**3as**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a yellow solid (90.7 mg, yield: 87%); mp 180−181 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.86 (d, J = 7.0 Hz, 2H), 7.60 (dd, J = 7.7, 1.7 Hz, 1H), 7.59−7.55 (m, 1H), 7.52 (d, J = 8.0 Hz, 1H), 7.43 (d, J = 7.7 Hz, 2H), 7.36−7.27 (m, 5H), 7.22−7.19 (m, 1H), 7.15 (t, J = 7.9 Hz, 2H), 7.01−6.94 (m, 3H), 6.87 (s, 1H), 4.65 (d, J = 9.8 Hz, 1H), 2.77−2.67 (m, 2H), 2.22 (d, J = 13.3 Hz, 1H), 2.15−1.97 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 166.6, 154.2, 142.0, 140., 133.6, 131.8, 130.5, 129.3, 129.1, 128.8, 128.8, 128.6, 128.3, 128.0, 127.7, 127.6, 127.4, 126.6, 126.0, 125.9, 123.9, 121.4, 83.7, 52.8, 27.8, 26.1; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{30}H_{24}BrN_3NaO$, 544.0995; found, 544.0997.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)-1-naphthamide (**3at**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (70.0 mg, yield: 71%); mp 188−189 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 8.17 (s, 1H), 7.88−7.78 (m, 5H), 7.70 (dd, J = 8.6, 1.7 Hz, 1H), 7.53−7.46 (m, 2H), 7.42−7.38 (m, 3H), 7.37−7.31 (m, 1H), 7.24−7.17 (m, 5H), 7.09 (s, 1H), 7.07−7.01 (m, 3H), 4.70 (dd, J = 8.4, 1.8 Hz, 1H), 2.78−2.65 (m, 2H), 2.27−2.20 (m, 1H), 2.12−2.05 (m, 1H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 166.7, 154.4, 142.0, 141.0, 134.9, 133.4, 132.5, 131.7, 131.0, 129.2, 129.1, 128.9, 128.8, 128.7, 128.3, 128.0, 128.0, 127.8, 127.7, 127.3, 127.0, 126.7, 126.0, 124.2, 123.2, 121.5, 83.3, 52.9, 27.9, 26.1; HRMS (ESI) m/z: [M + Na]$^+$ calcd. for $C_{34}H_{27}N_3NaO$, 516.2046; found, 516.2048.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9byl)cyclopropanecarboxamide (**3au**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (69.2 mg, yield: 85%); mp 150−151 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.84 (d, J = 7.5 Hz, 2H), 7.44 (t, J = 7.4 Hz, 2H), 7.39 (d, J = 7.2 Hz, 1H), 7.36−7.31 (m, 1H), 7.24−7.20 (m, 2H), 7.19−7.10 (m, 3H), 7.00−6.91 (m, 3H), 6.36 (s, 1H), 4.45 (d, J = 8.0 Hz, 1H), 2.67−2.55 (m, 2H), 2.15 (d, J = 13.1 Hz, 1H), 1.98−1.84 (m, 1H), 1.39−1.35 (m, 1H), 0.93 (s, 2H), 0.76−0.73 (m, 1H), 0.71−0.68 (m, 1H); ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 173.2, 153.8, 142.0, 140.9, 133.5, 131.8, 129.0, 128.8, 128.7, 128.0, 127.8, 127.1, 126.5, 125.9, 123.5, 120.9, 82.6, 52.9, 27.7, 26.0, 15.0, 7.7, 7.1; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{27}H_{26}N_3O$, 408.2070; found, 408.2069.

*N-((3a,9b)-1,3-Diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]-indazol-9b-yl)isobutyramide (**3av**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (74.5 mg, yield: 91%); mp 144−145 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.83 (d, J = 7.3 Hz, 2H), 7.43 (t, J = 7.4 Hz, 2H), 7.37 (t, J = 7.2 Hz, 1H), 7.28−7.24 (m, 1H), 7.23−7.19 (m, 2H), 7.16−7.12 (m, 3H), 6.98 (t, J = 7.3 Hz, 1H), 6.93 (d, J = 7.9 Hz, 2H), 6.23 (s, 1H), 4.45 (d, J = 7.3 Hz, 1H), 2.63−2.60 (m, 2H), 2.40−2.35 (m, 1H), 2.14 (d, J = 12.0 Hz, 1H), 1.99−1.91 (m, 1H), 1.14 (d, J = 6.9 Hz, 3H), 1.08 (d, J = 6.9 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 176.6, 153.9, 142.0, 140.9, 133.5, 131.9, 129.1, 128.8, 128.7, 128.1, 127.9, 127.0, 126.6, 125.9, 123.7, 121.1, 82.4, 52.9, 35.8, 27.8, 26.0, 19.7, 19.3; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{27}H_{28}N_3O$, 410.2227; found, 410.2227.

*N-((3a,9b)-5-Methyl-1,3-diphenyl-1,3a,4,5-tetrahydro-9bH-benzo[g]indazol-9b-yl)acetamide (**3aw**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 10:1 to 5:1) as a white solid (72.7 mg, yield: 92%); mp 167−168 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.84 (d, J = 7.4 Hz, 2H), 7.44 (t, J = 7.4 Hz, 2H), 7.40 (d, J = 7.1 Hz, 1H), 7.36−7.31 (m, 2H), 7.29−7.22 (m, 3H), 7.14 (t, J = 7.8 Hz, 2H), 6.96 (t, J = 7.3 Hz, 1H), 6.92 (d, J = 7.9 Hz, 2H), 6.16 (s, 1H), 4.49 (d, J = 7.7 Hz, 1H), 2.75 (s, 1H), 2.01 (d, J = 12.2 Hz, 1H), 1.97 (s, 3H), 1.77−1.68 (m, 1H), 1.29 (d, J = 6.7 Hz, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.0, 154.2, 144.5, 141.9, 132.9, 131.8, 129.1, 128.8, 128.8, 128.3, 127.2, 126.6, 125.6, 124.4, 123.4, 120.5, 82.9, 51.80, 34.46, 29.8, 24.0, 18.4; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{26}H_{26}N_3O$, 396.2070; found, 396.2070.

*N-(1,3,5-Triphenyl-4,5-dihydro-1H-pyrazol-5-yl)acetamide (**4a**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a yellow solid (60.4 mg, yield: 85%); mp 178−179 °C. ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.72 (d, J = 8.5 Hz, 2H), 7.49 (d, J = 7.5 Hz, 2H), 7.40−7.34 (m, 4H), 7.32 (q, J = 7.2 Hz, 2H), 7.13 (t, J = 7.9 Hz, 2H), 7.05 (d, J = 7.8 Hz, 2H), 6.82 (t, J = 7.2 Hz, 1H), 6.53 (s, 1H), 4.43 (d, J = 17.8 Hz, 1H), 3.42 (d, J = 17.8 Hz, 1H), 1.87 (s, 3H); ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 170.1, 147.4, 142.8, 142.0, 132.3, 129.2, 128.8, 128.8, 128.5, 128.3, 125.8, 125.4, 120.3, 115.5, 82.7, 50.3, 23.7; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{23}H_{22}N_3O$, 356.1757; found, 356.1756.

*N-(5-(4-Fluorophenyl)-1,3-diphenyl-4,5-dihydro-1H-pyrazol-5-yl)acetamide (**4b**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (65.7 mg, yield: 88%); mp 176−177 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.73−7.71 (m, 2H), 7.49−7.44 (m, 2H), 7.41−7.32 (m, 4H), 7.17−7.13 (m, 2H), 7.04 (d, J = 8.6 Hz, 3H), 6.85 (t, J = 7.3 Hz, 1H), 6.46 (s, 1H), 4.46 (d, J = 17.9 Hz, 1H), 3.44 (d, J = 17.9 Hz, 1H), 1.92 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.2, 162.4 (d, J = 248.0 Hz), 147.8, 141.9, 138.6 (d, J = 3.2 Hz), 132.2, 129.0, 128.9, 128.6, 127.9 (d, J = 8.2 Hz), 125.9, 120.7, 116.1 (d, J = 21.7 Hz), 115.7, 82.5, 50.4, 23.9; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{23}H_{21}FN_3O$, 374.1663; found, 374.1663.

*N-(5-(4-Chlorophenyl)-1,3-diphenyl-4,5-dihydro-1H-pyrazol-5-yl)acetamide (**4c**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (70.0 mg, yield: 90%); mp 174−176 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.73−7.71 (m, 2H), 7.44−7.41 (m, 2H), 7.40−7.33 (m, 5H), 7.17−7.14 (m, 2H), 7.03 (d, J = 7.8 Hz, 2H), 6.86 (t, J = 7.3 Hz, 1H), 6.41 (s, 1H), 4.46 (d, J = 18.0 Hz, 1H), 3.44 (d, J = 18.0 Hz, 1H), 1.92 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.2, 147.8, 141.8, 141.3, 134.2, 129.3, 129.1, 129.0, 128.6, 127.4, 127.2, 125.9, 120.8, 115.7, 82.5, 50.4, 23.9; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{23}H_{21}ClN_3O$, 390.1368; found, 390.1367.

*N-(5-(4-Bromophenyl)-1,3-diphenyl-4,5-dihydro-1H-pyrazol-5-yl)acetamide (**4d**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (75.4 mg, yield: 87%); mp 185−186 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.72 (d, J = 7.1 Hz, 2H), 7.50 (d, J = 8.6 Hz, 2H), 7.41−7.31 (m, 5H), 7.17−7.12 (m, 2H), 7.03 (d, J = 8.0 Hz, 2H), 6.86 (d, J = 7.3 Hz, 1H), 6.44 (s, 1H), 4.46 (d, J = 18.0 Hz, 1H), 3.43 (d, J = 18.0 Hz, 1H), 1.91 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.3, 147.8, 141.8, 141.8, 132.3, 132.2, 129.1, 129.0, 128.6, 127.6, 125.9, 122.4, 120.8, 115.7, 82.6, 50.4, 23.9; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{23}H_{21}BrN_3O$, 434.0863; found, 434.0866.

*N-(5-(3-Chlorophenyl)-1,3-diphenyl-4,5-dihydro-1H-pyrazol-5-yl)acetamide (**4e**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (68.5 mg, yield: 88%); mp 166−167 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.72−7.69 (m, 2H), 7.49 (s, 1H), 7.40−7.32 (m, 4H), 7.31−7.28 (m, 2H), 7.16−7.13 (m, 2H), 7.04−7.02 (m, 2H), 6.85 (t, J = 7.3 Hz, 1H), 6.59 (s, 1H), 4.41 (d, J = 17.9 Hz, 1H), 3.41 (d, J = 17.9 Hz, 1H), 1.86 (s, 3H); ^{13}C{^{1}H} NMR (125 MHz, $CDCl_3$): δ 170.4, 147.6, 145.0, 141.8, 135.2, 132.1, 130.6, 129.1, 128.9, 128.6, 128.6, 125.9, 125.9, 124.0, 120.7, 115.7, 82.4, 50.4, 23.8; HRMS (ESI) m/z: [M + H]$^+$ calcd. for $C_{23}H_{21}ClN_3O$, 390.1368; found, 390.1366.

*N-(5-(4-Methoxyphenyl)-1,3-diphenyl-4,5-dihydro-1H-pyrazol-5-yl)acetamide (**4f**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (69.3 mg, yield: 90%); mp 183−185 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.72 (d, J = 8.4 Hz, 2H), 7.42−7.29 (m, 5H), 7.14 (t, J

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= 7.9 Hz, 2H), 7.07 (d, *J* = 7.8 Hz, 2H), 6.89 (d, *J* = 8.8 Hz, 2H), 6.83 (t, *J* = 7.2 Hz, 1H), 6.47 (s, 1H), 4.41 (d, *J* = 17.8 Hz, 1H), 3.79 (s, 3H), 3.40 (d, *J* = 17.8 Hz, 1H), 1.87 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 170.1, 159.4, 147.5, 142.1, 135.0, 132.5, 128.8, 128.6, 127.4, 126.8, 125.9, 120.4, 115.6, 114.5, 82.6, 55.4, 50.3, 23.8; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{24}H_{24}N_3O_2$, 386.1863; found, 386.1863.

*N-(5-(4-(tert-Butyl)phenyl)-1,3-diphenyl-4,5-dihydro-1H-pyrazol-5-yl)acetamide (**4g**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (67.4 mg, yield: 82%); mp 170−171 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.73−7.70 (m, 2H), 7.43−7.29 (m, 7H), 7.18−7.12 (m, 2H), 7.09 (d, *J* = 8.6 Hz, 2H), 6.83 (t, *J* = 7.2 Hz, 1H), 6.44 (s, 1H), 4.42 (d, *J* = 17.7 Hz, 1H), 3.43 (s, 1H), 1.87 (s, 3H), 1.32 (s, 9H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 170.0, 151.4, 147.4, 142.2, 140.0, 132.5, 128.8, 128.76, 128.5, 126.2, 125.7, 125.1, 120.2, 115.6, 82.6, 50.2, 34.6, 31.3, 23.8; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{27}H_{30}N_3O$, 412.2383; found, 412.2383.

*N-(5-(4-Ethylphenyl)-1,3-diphenyl-4,5-dihydro-1H-pyrazol-5-yl)-acetamide (**4h**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (63.6 mg, yield: 83%); mp 188−189 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.72−7.70 (m, 2H), 7.42−7.29 (m, 5H), 7.21 (d, *J* = 8.3 Hz, 2H), 7.16−7.12 (m, 2H), 7.08−7.06 (m, 2H), 6.82 (t, *J* = 7.2 Hz, 1H), 6.49 (s, 1H), 4.41 (d, *J* = 17.7 Hz, 1H), 3.40 (d, *J* = 17.7 Hz, 1H), 2.65 (q, *J* = 7.5 Hz, 2H), 1.86 (s, 3H), 1.23 (t, *J* = 7.6 Hz, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 170.1, 147.4, 144.5, 142.2, 140.3, 132.5, 128.8, 128.7, 128.5, 126.1, 125.9, 125.4, 120.3, 115.6, 82.6, 50.3, 28.5, 23.8, 15.5; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{25}H_{26}N_3O$, 384.2070; found, 384.2071.

*N-(1,3-Diphenyl-5-(p-tolyl)-4,5-dihydro-1H-pyrazol-5-yl)-acetamide (**4i**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1 to 2:1) as a white solid (65.7 mg, yield: 89%); mp 177−178 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.72 (d, *J* = 7.3 Hz, 2H), 7.38−7.29 (m, 5H), 7.19 (d, *J* = 8.0 Hz, 2H), 7.14 (t, *J* = 7.8 Hz, 2H), 7.07 (d, *J* = 8.0 Hz, 2H), 6.82 (t, *J* = 7.2 Hz, 1H), 6.44 (s, 1H), 4.42 (d, *J* = 17.8 Hz, 1H), 3.40 (d, *J* = 17.8 Hz, 1H), 2.35 (s, 3H), 1.88 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 170.1, 147.4, 142.1, 140.1, 138.2, 132.5, 129.9, 128.8, 128.5, 125.9, 125.4, 120.3, 115.6, 82.6, 50.4, 23.8, 21.1; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{24}H_{24}N_3O$, 370.1914; found, 370.1914.

General Procedure for the One-Pot Synthesis of Pyrazoles 5. To a dried test tube with a magnetic stirring bar under N_2 at room temperature were added hydrazonoyl chloride 1 (0.3 mmol) and enamide 2 (0.20 mmol), followed by addition of K_2CO_3 (0.6 mmol). Then, dichloromethane (2.00 mL) was introduced by a syringe and the mixture was stirred at room temperature for 24 h. The reaction mixture was treated with $AlCl_3$ (0.02 mmol) and stirred at room temperature for 0.5 h. The reaction was then quenched with water and extracted with ethyl acetate three times. The combined organic layer was washed with brine, dried over Na_2SO_4, and the solvent was removed by rotary evaporation. The residue was purified by column chromatography on silica gel (petroleum/ethyl acetate = 20:1) to afford the desired pyrazoles **5**.

*5-(4-Fluorophenyl)-1,3-diphenyl-1H-pyrazole (**5a**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (52.1 mg, yield: 83%); mp 139−140 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.85−7.78 (m, 2H), 7.35−7.30 (m, 2H), 7.25−7.17 (m, 6H), 7.15−7.11 (m, 2H), 6.93−6.86 (m, 2H), 6.68 (s, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 161.5 (d, *J* = 248.7 Hz), 150.9, 142.3, 138.9, 131.9, 129.5 (d, *J* = 8.3 Hz), 127.9, 127.6, 127.0, 126.5, 125.6 (d, *J* = 3.3 Hz), 124.7, 124.2, 114.6 (d, *J* = 21.8 Hz), 104.1; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{21}H_{16}FN_2$, 315.1292; found, 315.1290.

*5-(4-Chlorophenyl)-1,3-diphenyl-1H-pyrazole (**5b**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (56.1 mg, yield: 85%); mp 135−136 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.95−7.86 (m, 2H), 7.45−7.39 (m, 2H), 7.37−7.26 (m, 8H), 7.21−7.16 (m, 2H), 6.80 (s, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$); δ 152.1, 143.2, 139.9, 134.4, 132.9, 130.0, 129.1, 129.05, 128.8, 128.7, 128.2, 127.7, 125.9, 125.4, 105.3; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{21}H_{16}ClN_2$, 331.0997; found, 331.0996.

*5-(4-Bromophenyl)-1,3-diphenyl-1H-pyrazole (**5c**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a yellow solid (65.8 mg, yield: 88%); mp 120−121 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.93−7.88 (m, 2H), 7.43−7.40 (m, 4H), 7.36−7.27 (m, 6H), 7.14−7.08 (m, 2H), 6.79 (s, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 152.2, 143.2, 139.9, 132.9, 131.8, 130.3, 129.5, 129.15, 128.8, 128.2, 127.8, 125.9, 125.4, 122.7, 105.4; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{21}H_{16}BrN_2$, 375.0491; found, 375.0493.

*5-(3-Chlorophenyl)-1,3-diphenyl-1H-pyrazole (**5d**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (55.5 mg, yield: 84%); mp 144−145 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.92−7.90 (m, 2H), 7.44−7.39 (m, 2H), 7.38−7.26 (m, 8H), 7.19 (t, *J* = 7.9 Hz, 1H), 7.07−7.05 (m, 1H), 6.82 (s, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 152.1, 142.9, 139.9, 134.47, 132.9, 132.3, 129.8, 129.1, 128.8, 128.7, 128.4, 128.2, 127.8, 127.0, 125.9, 125.4, 105.6; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{21}H_{16}ClN_2$, 331.0997; found, 331.0996.

*1,3,5-Triphenyl-1H-pyrazole (**5e**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a yellow solid (51.5 mg, yield: 87%); mp 142−143 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.99−7.87 (m, 2H), 7.43 (t, *J* = 7.6 Hz, 2H), 7.39−7.27 (m, 11H), 6.83 (s, 1H); ^{13}C NMR (125 MHz, $CDCl_3$): δ 152.0, 144.4, 140.1, 133.0, 130.6, 128.9, 128.8, 128.7, 128.5, 128.3, 128.0, 127.4, 125.8, 125.3, 105.2; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{21}H_{17}N_2$, 297.1386; found, 297.1385.

*1,3-Diphenyl-5-(p-tolyl)-1H-pyrazole (**5f**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (52.7 mg, yield: 85%); mp 133−134 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 8.00−7.88 (m, 2H), 7.44−7.27 (m, 8H), 7.18−7.09 (m, 4H), 6.78 (s, 1H), 2.34 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 152.0, 144.5, 140.3, 138.3, 133.2, 129.3, 128.9, 128.7, 128.7, 128.0, 127.7, 127.4, 125.9, 125.4, 105.0, 21.3; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{22}H_{19}N_2$, 311.1543; found, 311.1541.

*5-(4-Methoxyphenyl)-1,3-diphenyl-1H-pyrazole (**5g**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (59.4 mg, yield: 91%); mp 129−130 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.97−7.88 (m, 2H), 7.44−7.27 (m, 8H), 7.21−7.16 (m, 2H), 6.85−6.81 (m, 2H), 6.76 (s, 1H), 3.78 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 159.6, 151.9, 144.3, 140.3, 133.2, 130.1, 129.0, 128.7, 128.0, 127.4, 125.9, 125.4, 123.0, 114.0, 104.8, 55.3; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{22}H_{19}N_2O$, 327.1492; found, 327.1490.

*5-(4-(tert-Butyl)phenyl)-1,3-diphenyl-1H-pyrazole (**5h**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (61.3 mg, yield: 87%); mp 122−123 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.94−7.90 (m, 2H), 7.42−7.24 (m, 10H), 7.21−7.17 (m, 2H), 6.79 (s, 1H), 1.29 (s, 9H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 152.0, 151.5, 144.5, 140.4, 133.2, 129.0, 128.7, 128.4, 128.1, 127.7, 127.5, 125.9, 125.5, 125.5, 105.1, 34.7, 31.3; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{25}H_{25}N_2$, 353.2012; found, 353.2011.

*5-(4-Ethylphenyl)-1,3-diphenyl-1H-pyrazole (**5i**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (54.5 mg, yield: 84%); mp 140−141 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.96−7.89 (m, 2H), 7.44−7.24 (m, 8H), 7.20−7.10 (m, 4H), 6.78 (s, 1H), 2.63 (q, *J* = 7.6 Hz, 2H), 1.22 (t, *J* = 7.6 Hz, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 152.0, 144.6, 140.3, 133.2, 129.0, 128.7, 128.7, 128.2, 128.1, 128.0, 127.9, 127.4, 125.9, 125.4, 105.1, 28.7, 15.4; HRMS (ESI) *m*/*z*: $[M + H]^+$ calcd. for $C_{23}H_{21}N_2$, 325.1699; found, 325.1698.

*3-(4-Fluorophenyl)-1-phenyl-4,5-dihydro-1H-benzo[g]indazole (**5j**).* The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a yellow solid (60.5 mg, yield: 89%); mp 200−201 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.78−7.71 (m, 2H), 7.56 (d, *J* = 7.3 Hz, 2H), 7.53−7.41 (m, 3H), 7.31

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(d, J = 7.4 Hz, 1H), 7.19−7.09 (m, 3H), 7.01 (t, J = 7.5 Hz, 1H), 6.82 (d, J = 7.8 Hz, 1H), 3.03−3.00 (m, 2H), 2.96−2.92 (m, 2H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 162.5 (d, J = 246.6 Hz), 147.8, 140.8, 139.1, 137.2, 129.6 (d, J = 3.2 Hz), 129.4, 129.0 (d, J = 8.2 Hz), 128.6, 128.4, 127.6, 126.9, 126.3, 125.9, 123.2, 117.3, 115.5 (d, J = 21.3 Hz), 30.8, 20.7; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{18}FN_2$, 341.1449; found, 341.1447.

3-(4-Chlorophenyl)-1-phenyl-4,5-dihydro-1H-benzo[g]indazole (**5k**). The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (64.8 mg, yield: 91%); mp 188−189 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.74−7.69 (m, 2H), 7.57−7.53 (m, 2H), 7.51−7.38 (m, 5H), 7.30 (d, J = 7.3 Hz, 1H), 7.17−7.14 (m, 1H), 7.00 (t, J = 8.1 Hz, 1H), 6.81 (d, J = 7.7 Hz, 1H), 3.03−2.99 (m, 2H), 2.96−2.91 (m, 2H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 147.5, 140.8, 139.2, 137.1, 133.6, 132.0, 129.4, 128.8, 128.6, 128.6, 128.4, 127.6, 126.9, 126.4, 125.9, 123.2, 117.5, 30.7, 20.8; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{18}ClN_2$, 357.1153; found, 357.1153.

3-(4-Bromophenyl)-1-phenyl-4,5-dihydro-1H-benzo[g]indazole (**5l**). The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (73.6 mg, yield: 92%); mp 178−179 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.68−7.63 (m, 2H), 7.57−7.54 (m, 4H), 7.52−7.43 (m, 3H), 7.30 (d, J = 7.2 Hz, 1H), 7.18−7.145 (m, 1H), 7.00 (t, J = 8.2 Hz, 1H), 6.81 (d, J = 7.8 Hz, 1H), 3.03−2.99 (m, 2H), 2.97−2.92 (m, 2H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 147.5, 140.8, 139.3, 137.1, 132.5, 131.7, 129.4, 128.9, 128.6, 128.5, 127.6, 126.8, 126.4, 125.9, 123.2, 121.8, 117.5, 30.7, 20.8; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{18}BrN_2$, 401.0648; found, 401.0649.

1-Phenyl-3-(p-tolyl)-4,5-dihydro-1H-benzo[g]indazole (**5m**). The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (57.1 mg, yield: 85%); mp 154−155 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.68−7.66 (m, 2H), 7.59−7.55 (m, 2H), 7.52−7.42 (m, 3H), 7.31 (d, J = 7.0 Hz, 1H), 7.27−7.25 (m, 2H), 7.18-7.14 (m, 1H), 7.00 (t, J = 7.6 Hz, 1H), 6.83 (d, J = 7.7 Hz, 1H), 3.05−3.00 (m, 2H), 2.99−2.93 (m, 2H), 2.40 (s, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 148.7, 140.9, 138.9, 137.5, 137.3, 130.6, 129.3, 129.3, 128.5, 128.2, 127.4, 127.3, 127.1, 126.3, 125.9, 123.2, 117.5, 30.8, 21.3, 20.8; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{24}H_{21}N_2$, 337.1699; found, 337.1697.

Ethyl 1-Phenyl-4,5-dihydro-1H-benzo[g]indazole-3-carboxylate (**5n**). The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a yellow solid (55.4 mg, yield: 87%); mp 152−153 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.56−7.47 (m, 5H), 7.30 (d, J = 7.4 Hz, 1H), 7.18−7.15 (m, 1H), 6.98 (t, J = 7.6 Hz, 1H), 6.73 (d, J = 7.7 Hz, 1H), 4.45 (q, J = 7.1 Hz, 2H), 3.13−3.07 (m, 2H), 3.05−2.97 (m, 2H), 1.43 (t, J = 7.1 Hz, 3H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 163.0, 140.4, 140.2, 139.6, 137.4, 129.4, 129.1, 128.7, 128.0, 126.4, 126.2, 126.2, 123.0, 122.9, 60.9, 30.2, 20.2, 14.5; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{20}H_{19}N_2O_2$, 319.1441; found, 319.1441.

1,3-Diphenyl-4,5-dihydro-1H-benzo[g]indazole (**5o**). The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a yellow solid (54.8 mg, yield: 85%); mp 176−177 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.81−7.75 (m, 2H), 7.59−7.56 (m, 2H), 7.51−7.47 (m, 2H), 7.45 (t, J = 7.6 Hz, 3H), 7.35 (t, J = 7.4 Hz, 1H), 7.31 (d, J = 7.5 Hz, 1H), 7.18−7.15 (m, 1H), 7.01 (t, J = 8.2 Hz, 1H), 6.83 (d, J = 8.3 Hz, 1H), 3.05−3.00 (m, 2H), 3.00−2.95 (m, 2H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 148.6, 140.9, 139.0, 137.2, 133.4, 129.3, 128.6, 128.5, 128.3, 127.7, 127.5, 127.4, 127.0, 126.3, 125.9, 123.1, 117.6, 30.8, 20.8; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{19}N_2$, 323.1543; found, 323.1543.

3-Phenyl-1-(p-tolyl)-4,5-dihydro-1H-benzo[g]indazole (**5p**). The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (63.9 mg, yield: 95%); mp 150−151 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.75−7.65 (m, 2H), 7.38−7.31 (m, 4H), 7.27−7.23 (m, 1H), 7.19 (t, J = 6.7 Hz, 3H), 7.06−7.03 (m, 1H), 6.91 (t, J = 8.1 Hz, 1H), 6.76 (d, J = 7.3 Hz, 1H), 2.93−2.89 (m, 2H), 2.88−2.84 (m, 2H), 2.34 (s, 3′H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 147.3, 137.9, 137.4, 137.2, 136.1, 132.5, 127.5, 127.4, 126.6, 126.3, 126.3, 126.0, 125.2, 124.7, 122.0, 116.2, 29.7, 20.2, 19.7; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{24}H_{21}N_2$, 337.1699; found, 337.1697.

1-(4-Chlorophenyl)-3-phenyl-4,5-dihydro-1H-benzo[g]indazole (**5q**). The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 20:1) as a white solid (62.0 mg, yield: 87%); mp 177−178 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.76−7.74 (m, 2H), 7.54−7.50 (m, 2H), 7.47−7.42 (m, 4H), 7.39−7.34 (m, 1H), 7.32 (d, J = 7.4 Hz, 1H), 7.20−7.17 (m, 1H), 7.05 (t, J = 8.1 Hz, 1H), 6.87 (d, J = 7.7 Hz, 1H), 3.03−2.99 (m, 2H), 2.98−2.92 (m, 2H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 149.1, 139.4, 139.1, 137.3, 133.9, 133.2, 129.5, 128.7, 127.9, 127.7, 127.4, 127.0, 126.8, 126.4, 123.1, 118.0, 30.8, 20.8; HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{18}ClN_2$, 357.1153; found, 357.1153.

Gram-Scale Synthesis. To a dried test tube with a magnetic stirring bar under N_2 at room temperature were added hydrazonoyl chloride **1a** (4.65 mmol, 1.5 equiv) and enamide **2r** (3.1 mmol, 1 equiv), followed by the addition of K_2CO_3 (1.28 g, 9.3 mmol). Then, dichloromethane (31 mL) was introduced and the mixture was stirred at room temperature for 24 h. The solvent was removed by rotary evaporation, and the residue was purified by column chromatography on silica gel (petroleum/ethyl acetate = 10:1 to 5:1) to afford the desired product **3ar** (1.29 g, 80% yield).

Procedure for the Derivation. To a solution of **3ar** (200 mg, 0.385 mmol) in toluene (10 mL) under a nitrogen atmosphere was added $Pd(P(Ph)_3)_4$ (45 mg, 0.039 mmol). After stirring for 30 min, a solution of phenylboronic acid in ethanol (5 mL) and saturated $NaHCO_3$ (3 mL) was added. Then, the mixture was heated to reflux in an oil bath for 24 h. After cooling to room temperature, the mixture was added to saturated aqueous NaCl solution and extracted with ethyl acetate. The organic layer was combined, washed with saturated brine, dried with anhydrous Na_2SO_4, and concentrated. The residue was chromatographed on silica gel to give the desired product **6**. The product was obtained by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1) as a yellow solid (102 mg, yield: 51%); mp 110−111 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.85 (d, J = 7.2 Hz, 2H), 7.73 (d, J = 8.4 Hz, 2H), 7.61 (d, J = 8.4 Hz, 2H), 7.56 (d, J = 7.3 Hz, 2H), 7.45−7.39 (m, 4H), 7.38−7.33 (m, 3H), 7.24−7.17 (m, 5H), 7.03 (dd, J = 14.4, 7.4 Hz, 3H), 6.95 (s, 1H), 4.66 (d, J = 9.5 Hz, 1H), 2.82−2.67 (m, 2H), 2.23 (d, J = 13.3 Hz, 1H), 2.14−1.98 (m, 1H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 166.4, 154.4, 144.9, 142.0, 141.0, 139.9, 133.5, 132.5, 131.8, 129.2, 129.0, 128.9, 128.9, 128.3, 128.2, 128.1, 127.5, 127.5, 127.3, 126.7, 126.1, 125.9, 124.2, 121.6, 83.3, 52.9, 27.9, 26.1; HRMS (ESI) m/z: $[M + Na]^+$ calcd. for $C_{36}H_{29}N_3NaO$, 542.2203; found, 542.2204.

To a mixture of the ester **5n** (0.31 mmol, 0.1 g, 1.0 equiv) in methanol (5.0 mL) was added potassium hydroxide (0.62 mmol, 2.0 equiv). The resulting mixture was heated under reflux in an oil bath for 1 h. The mixture was allowed to cool to room temperature and then poured into water, acidified with 1 N hydrochloric acid, and extracted with ethyl acetate. The organic layer was combined, washed with saturated brine, dried with anhydrous Na_2SO_4, and concentrated to yield the acid. The crude product was used for next step without purification. A 25 mL round-bottom flask was charged with acid (0.31 mmol, 1.0 equiv), dry DCM (10 mL), and a catalytic amount of DMF. The reaction mixture was cooled to 0 °C and stirred for 5 min. Then, oxalyl chloride (0.47 mmol, 40 μL, 1.5 equiv) was added dropwise to the reaction mixture and stirred at room temperature for 2 h. The resulting mixture was concentrated under reduced pressure to afford the crude product and used directly for the next step. The white solid was dissolved in CH_2Cl_2 (2 mL). To the resulting solution were added 1-aminopiperidine (0.46 mmol, 1.5 equiv) and Et_3N (0.46 mmol, 1.5 equiv) at 0 °C. The mixture was warmed to room temperature and stirred for 1 h. Then, the mixture was quenched with brine and extracted with CH_2Cl_2. The combined organic layer was washed with brine, dried over Na_2SO_4, and the solvent was removed by rotary evaporation. The residue was purified by column chromatography on silica gel (petroleum/ethyl acetate = 10:1) to afford the product **7** as a white solid (92.1 mg, 79% yield over 3 steps); mp 195−196 °C. ^1H NMR (500 MHz, $CDCl_3$): δ 7.70 (s, 1H), 7.57−7.47 (m, 5H), 7.29 (d,

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J = 7.4 Hz, 1H), 7.16 (t, J = 7.5 Hz, 1H), 6.97 (t, J = 7.7 Hz, 1H), 6.72 (d, J = 7.6 Hz, 1H), 3.17−3.12 (m, 2H), 3.04−2.96 (m, 2H), 2.88−2.84 (m, 3H), 1.90−1.70 (m, 5H), 1.53−1.38 (m, 2H); $^{13}C\{^1H\}$ NMR (125 MHz, $CDCl_3$): δ 160.0, 141.8, 140.4, 139.7, 137.7, 129.5, 129.0, 128.8, 127.9, 126.2, 126.1, 125.9, 122.8, 122.1, 57.2, 30.2, 25.4, 23.4, 19.7. HRMS (ESI) m/z: $[M + H]^+$ calcd. for $C_{23}H_{25}N_4O$, 373.2023; found, 373.2020.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.joc.0c02244.

Copies of ^1H and ^{13}C NMR spectra for all compounds (PDF)

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Notes

The authors declare no competing financial interest.

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