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CuI-Catalyzed Amination of Arylhalides with Guanidines or Amidines: A Facile Synthesis of 1-*H*-2-Substituted Benzimidazoles

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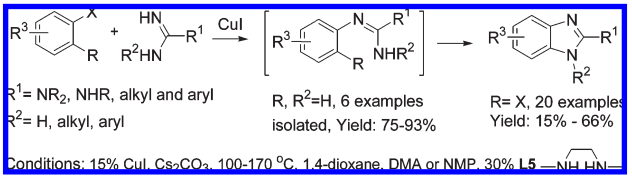

CuI/**L5** (*N*,*N*′-dimethylethylenediamine) proves to be an efficient catalyst system for the amination of arylhalides with guanidines. The same catalyst system is then successfully applied to the one-step synthesis of 1-*H*-2-amino-benzimidazoles through tandem aminations of 1,2-dihaloarenes in modest yields. This methodology is also applicable for the preparation of 1-*H* or 1-substutituted 2-aryl- or 2-alkyl-benzimidazoles.

Benzimidazoles, a "privileged" structure, is a hugely important class of compounds for the pharmaceutical industry.[1] The benzimidazole core structure can be found in many commercial drugs such as Prilosec, Nexium, Protonix, Atacand, Famvir, and Vermox, as well as numerous experimental drug candidates in a wide range of therapeutic areas.[2] Therefore, it is not surprising that the synthesis of benzimidazoles has always been of great interest to organic chemists.[3]

During our drug discovery efforts, we became interested in the facile assembly of 1-*H*-2-amino-benzimidazoles, molecules that have exhibited antihistamine,[4] immunosuppressive,[5] anti-inflammatory,[6] analgesic,[7] antiviral,[8] and antibacterial[9] activities. The prevailing synthetic strategy for this class of compounds is through a three-step sequence, starting from a 1,2-diaminobenzene (Scheme 1). 1,3-Dihydro-benzoimidazol-2-one formation is followed by activation of the ketone to chloride or sulfur derivatives[10] and then the displacement with an amine to provide the desired 1-*H*-2-amino-benzimidazole. Depending on the nucleophilicity of the amine, protection of the NH proton is often required prior to the amination step, which adds two steps to the overall sequence. In addition, 1,2-diaminobenzenes with the desired substitution pattern are often unstable and difficult to prepare. Apparently, there is a need for a more concise synthesis. Herein, we report our results on a one-step synthesis of 1-*H*-2-amino-benzimidazoles through tandem amination of 1,2-dihalobenzenes with substituted guanidines. This methodology is also quite general in preparing other 2-substituted benzimidazoles.

We envisioned that a direct metal-catalyzed double amination of 1,2-dihalobenzenes with substituted guanidines might offer one-step access to 1-*H*-2-amino-benzimidazole (Scheme 2). In pursuit of this goal, we recognized two potential obstacles. First, despite extensive studies on the metal-catalyzed Buchwald−Hartwig amination reaction,[11] amination of arylhalides with guanidines has rarely been found in the literature. Although a few intramolecular cases exist,[12] to the best of our knowledge, the intermolecular catalytic amination of nonactivated arylhalides with guanidines has not been reported.[13] The relative paucity of examples might result from the proclivity of guanidine to strongly bind many transition metals.[14] Alternatively, guanidines or

(1) Horton, D. A.; Bourne, G. T.; Smythe, M. L. *Chem. Rev.* **2003**, *103*, 893–930.

(2) Alamgir, M.; Black, D. St. C.; Kumar, N. Synthesis, reactivity and biological activity of benzimidazoles. In *Topics in Heterocyclic Chemistry*; Springer: Berlin, Heidelberg, 2007; Vol. 9, pp 87−118 and references therein.

(3) For recent examples, see: (a) Brasche, G.; Buchwald, S. L. *Angew. Chem., Int. Ed.* **2008**, *47*, 1932–1934. (b) Shen, M.; Driver, T. G. *Org. Lett.* **2008**, *10*, 3367–3370. (c) Bahrami, K.; Khodaei, M. M.; Naali, F. *J. Org. Chem.* **2008**, *73*, 6835–6837. (d) Goossen, L. J.; Knauber, T. *J. Org. Chem.* **2008**, *73*, 8631–8634. (e) Yamamoto, Y.; Mizuno, H.; Tsuritani, T.; Mase, T. *J. Org. Chem.* **2009**, *74*, 1394–1396. (f) Coffinier, D.; El Kaim, L.; Grimaud, L. *Org. Lett.* **2009**, *11*, 995–997.

(4) (a) Lemura, R.; Kawashima, T.; Fukuda, T.; Ito, K.; Tsukamoto, G. *J. Med. Chem.* **1986**, *29*, 1178–1183. (b) Lorenzi, S.; Mor, M.; Bordi, F.; Rivara, S.; Rivara, M.; Morini, G.; Bertoni, S.; Ballabeni, V.; Barocelli, E.; Plazzi, P. V. *Bioorg. Med. Chem.* **2005**, *13*, 5647–5657.

(5) Zhang, G.; Ren, P.; Gray, N. S.; Sim, T.; Liu, Y.; Wang, X.; Che, J.; Tian, S.; Sandberg, M. L.; Spalding, T. A.; Romeo, R.; Iskandar, M.; Chow, D.; Deidel, H. M.; Karanewsky, D. S.; He, Y. *Bioorg. Med. Chem. Lett.* **2008**, *18*, 5618–5621.

(6) Blythin, D. J.; Kaminski, J. J.; Domalski, M. S.; Spitler, J.; Solomon, D. M.; Conn, D. J.; Wong, S. C.; Verbiar, L. L.; Bober, L. A.; Chiu, P. J. S.; Watnick, A. S.; Siegel, M. I.; Hilbert, J. M.; McPhail, A. T. *J. Med. Chem.* **1986**, *29*, 1099–1113.

(7) (a) Shao, B.; Huang, J.; Sun, Q.; Valenzani, K. J.; Schmid, L.; Nolan, S. *Bioorg. Med. Chem. Lett.* **2005**, *15*, 719–723. (b) Ognyanov, V. I.; Balan, C.; Bannon, A. W.; Bo, Y.; Dominguez, C.; Fotsch, C.; Gore, V. K.; Klionsky, L.; Ma, V. V.; Qian, Y.; Tamir, R.; Wang, X.; Xi, N.; Xu, S.; Zhu, D.; Gavva, N. R.; Treanor, J. J. S.; Norman, M. H. *J. Med. Chem.* **2006**, *49*, 3719–3742.

(8) Bonfanti, J-F; Meyer, C.; Doublet, F.; Fortin, J.; Muller, P.; Queguiner, L.; Gevers, T.; Janssens, P.; Szel, H.; Willebrords, R.; Timmerman, P.; Wuyts, K.; Van Remoortere, P.; Janssens, F.; Wigerinck, P.; Andries, K. *J. Med. Chem.* **2008**, *51*, 875–896.

(9) Ozden, S.; Atabey, D.; Yildiz, S.; Goker, H. *Eur. J. Med. Chem.* **2008**, *43*, 1390–1402.

(10) Mavrova, A. T.; Denkova, P.; Tsenov, Y. A.; Anichina, K. K.; Vutchev, D. I. *Bioorg. Med. Chem.* **2007**, *15*, 6291–6297. (b) Lan, P.; Romero, F. A.; Malcolm, T. S.; Stevens, B. D.; Wodka, D.; Makara, G. M. *Tetrahedron Lett.* **2008**, *49*, 1910–1914.

(11) (a) Surry, D. S.; Buchwald, S. L. *Angew. Chem., Int. Ed.* **2008**, *47*, 6338–6361. (b) Hartwig, J. F. *Acc. Chem. Res.* **2008**, *41*, 1534–1544.

(12) (a) Evindar, G.; Batey, R. A. *Org. Lett.* **2003**, *5*, 133–136. (b) Szczepankiewicz, B. G.; Rohde, J. J.; Kurukulasuriyz, R. *Org. Lett.* **2005**, *7*, 1833–1835.

(13) A recent paper described the synthesis of quinazolinones from 2-halobenzoic acid and amidine/guanidine, presumably through an amination-cyclization sequence. However, the amination intermediate was not isolated. Liu, X.; Fu, H.; Jiang, Y.; Zhao, Y. *Angew. Chem., Int. Ed.* **2009**, *48*, 348–351.

(14) Oakley, S. H.; Soria, D; Coles, B.; Hitchcock, M. P. *Dalton Trans.* **2004**, 537–546.

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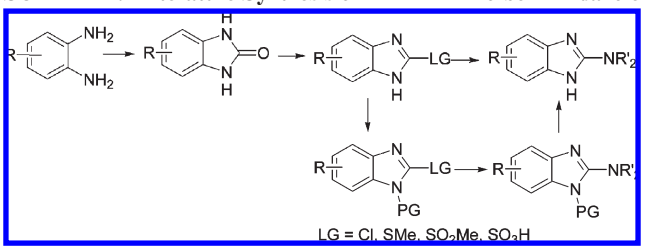
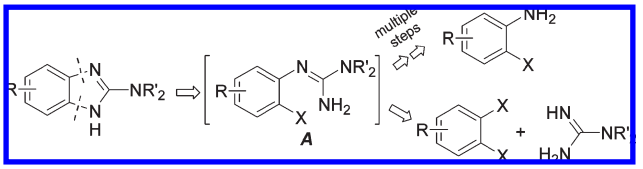
SCHEME 1. Literature Synthesis of 1-*H*-2-Amino-benzimidazole



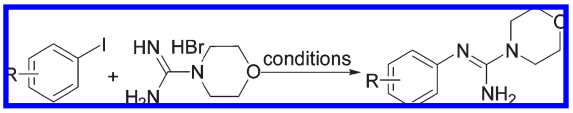
SCHEME 2. One-Step Synthesis of 1-*H*-2-Amino-benzimidazole



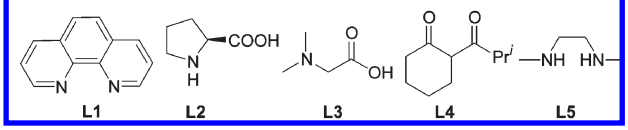
amidines might not be stable enough to survive certain metal-catalyzed reaction conditions. In fact, alkyl and aryl amidines have been used as ammonia surrogates in the CuI-catalyzed synthesis of primary anilines from arylhalides.[15] After the initial coupling, an intramolecular amination is still required to complete the benzimidazole construction. Although metal-catalyzed intramolecular aryl guanidinylation through an isolated intermediate **A** (Scheme 2)[12] or amidinylation[16] have been utilized to construct benzimidazoles, almost all of the prior art has involved *N*-substituted benzimidazoles. In our case, the unsubstituted NH_2 group of intermediate **A** presents a unique challenge because of its basicity and the potential to coordinate with the metal catalyst. In addition, it was not assured that a single catalyst system would be suitable for the two tandem amination steps. Nevertheless, we felt that a suitable metal catalyst system had a good chance to effect this novel transformation.

To study the intermolecular amination reaction of arylhalides with guanidines, we first investigated the reaction between 3,4-dichloro-iodobenzene and morpholine-4-carboxamidine (Table 1). Our initial attempts with Pd-based amination conditions[17] failed to produce any of the desired amination product (entries 1 and 2). We quickly turned our attention to Cu-based amination conditions with five representative ligands, i.e., 1,10-phenanthroline **L1** (entry 3),[18] amino acids **L2** and **L3** (entries 4 and 5),[19] diketone **L4** (entry 6),[20] and

TABLE 1. Optimization of Amination Reaction Conditions



entry	R	conditions	product	yield[b]
1	*m*, *p*-di-Cl	$Pd_2(dba)_3$, *t*-Bu-XPhos, $NaOBu^t$ toluene, Microwave, 200 °C, 10 min	**1**	0
2	*m*, *p*-di-Cl	$Pd_2(dba)_3$, S-Phos, Cs_2CO_3 NMP, Microwave, 200 °C, 10 min	**1**	0
3	*m*, *p*-di-Cl	CuI, **L1**, Cs_2CO_3, NMP, 120 °C, 24 h	**1**	0
4	*m*, *p*-di-Cl	CuI, **L2**, Cs_2CO_3, NMP, 120 °C, 24 h	**1**	60%
5	*m*, *p*-di-Cl	CuI, **L3**, Cs_2CO_3, DMSO, 120 °C, 24 h	**1**	40%[a]
6	*m*, *p*-di-Cl	CuI, **L4**, Cs_2CO_3, DMF, 120 °C, 24 h	**1**	35%[a]
7	*m*, *p*-di-Cl	10% CuI, 20% **L5**, 3 eq Cs_2CO_3 1,4-dioxane, 100 °C, 24 h	**1**	86%
8	*p*-Me	same as above	**2**	89%
9	*p*-NO_2	same as above	**3**	93%
10	*p*-COOMe	same as above	**4**	83%
11	*p*-MeO	same as above	**5**	75%
12	*p*-CN	same as above	**6**	84%
13		same as above	**2**	35%[a]



L1 **L2** **L3** **L4** **L5**

[a] Conversion based on HPLC analysis. SM remained. [b] Isolated yield.

diamine **L5** (entry 7).[21] Whereas the CuI/**L1** system was ineffective, CuI/**L2**−**L4** all afforded the desired amination product **1** in 35−60% yields. The best result we obtained was with the CuI/**L5** system, where an excellent 86% isolated yield was achieved under relatively mild conditions (100 °C in 1,4-dioxane). It is noteworthy that the reaction profile is rather clean and that compound **1** was isolated in >90% purity after simple aqueous workup without further purification (entry 7). We then investigated a number of aryliodides bearing various functional groups. Similarly, desired products **2**−**6** were isolated in good yield and purity after a simple aqueous workup procedure (entry 8−12). In comparison, when the same CuI/**L5** amination conditions were applied to 4-bromotoluene, the reaction was significantly slower with only 35% conversion after 24 h at 100 °C (entry 13).

Surprisingly, when the optimal CuI/**L5**/1,4-dioxane conditions were applied to the tandem double amination of 1,2-diiodobenzene in the attempt to prepare 1-*H*-2-aminobenzimidazole **7**, the reaction was incomplete after 24 h at 100 °C in 1,4-dioxane at only 30% conversion. The sluggishness of this reaction likely resulted from steric hindrance of the adjacent iodide (Table 2, entry 1). Simply switching the solvent to DMA and raising the reaction temperature to 150 °C provided desired compound **7** in decent

(15) (a) Gao, X.; Fu, H.; Qiao, R.; Jiang, Y.; Zhao, Y. *J. Org. Chem.* **2008**, *73*, 6864–6866. (b) Yang, D.; Fu., H.; Hu, L.; Jiang, Y.; Zhao, Y. *J. Org. Chem.* **2008**, *73*, 7841–7844.

(16) (a) Brain, C. T.; Brunton, S. A. *Tetrahedron Lett.* **2002**, *43*, 1893–1895. (b) Brain, C. T.; Steer, J. T. *J. Org. Chem.* **2003**, *68*, 6814–6816. (c) Alen, J.; Robeyns, K.; De Borggraeve, W. M.; Van Meervelt, L.; Compernolle, F. *Tetrahedron* **2008**, *64*, 8128–8133. (d) Li, Z.; Sun, H.; Jiang, H.; Liu, H. *Org. Lett.* **2008**, *10*, 3263–3266. (e) Viire, R. D.; Evindar, G.; Batey, R. A. *J. Org. Chem.* **2008**, *73*, 3452–3459. (f) Murru, S.; Patel, B. K.; Le Bras, J.; Muzart, J. *J. Org. Chem.* **2009**, *74*, 2217–2220.

(17) (a) Anderson, K. W.; Tundel, R. E.; Ikawa, T.; Atlman, R. A.; Buchwald, S. L. *Angew. Chem., Int. Ed.* **2006**, *45*, 6523–6527. (b) Zhen, N.; Anderson, K. W.; Huang, X.; Nguyen, H. N.; Buchwald, S. L. *Angew. Chem., Int. Ed.* **2007**, *46*, 7509–7512.

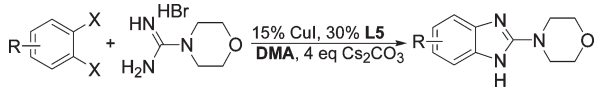
(18) Altman, R. A.; Koval, E. D.; Buchwald, S. L. *J. Org. Chem.* **2007**, *72*, 6190–6199.

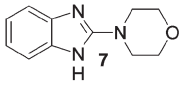
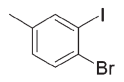
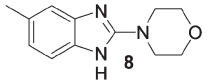
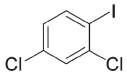
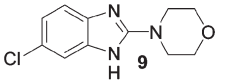
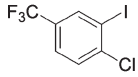
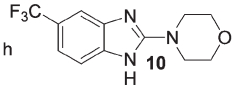
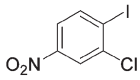
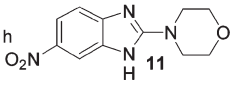
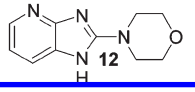
(19) (a) Zhang, H.; Cai, Q.; Ma. D. *J. Org. Chem.* **2005**, *70*, 5164–5173. (b) Ma, D.; Cai, Q. *Acc. Chem. Res.* **2008**, *41*, 1450–1460.

(20) Shafir, A.; Lichtor, P. A.; Buchwald, S. L. *J. Am. Chem. Soc.* **2007**, *129*, 3490–3491.

(21) (a) Jiang, L.; Job, G. E.; Klapars, A.; Buchwald, S. L. *Org. Lett.* **2003**, *5*, 3667–3669. (b) Martin, R.; Larsen, C. H.; Cuenca, A.; Buchwald, S. L. *Org. Lett.* **2007**, *9*, 3379–3382.

TABLE 2. Benzimidazole Synthesis with 1,2-Dihalobenzenes

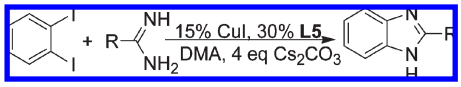


entry	substrate	conditions	product	yield[b]
1	1,2-di-iodo-benzene	100 °C, 24 h[c]	 **7**	(30%)[a]
2	1,2-di-iodo-benzene	150 °C, 16 h	**7**	52% (77%)[a]
3	1-Br-2-I-benzene	150 °C, 16 h	**7**	(76%)[a]
4	1-Cl-2-I-benzene	150 °C, 16 h	**7**	(25%)[a]
5	1,2-dibromo-benzene	150 °C, 16 h	**7** + 25% sm	(40%)[a]
6	1-Br-2-Cl-benzene	150 °C, 16 h	**7** + 28% sm	(12%)[a]
7	1,2-dichloro-benzene	150 °C, 16 h	**7**	0
8		165 °C, 16 h	 **8**	53%
9		165 °C, 16 h	 **9**	26%
10		165 °C, 16 h	 **10**	18%
11		150 °C, 16 h	 **11**	15%
12		150 °C, 16 h	 **12**	16%

[a] HPLC yield with internal standard. [b] Isolated yield. [c] 1,4-dioxane as the solvent.

yield (entry 2, 77% HPLC assay yield and 52% isolated yield[22]). We then explored other 1,2-dihalobenzenes as the coupling partners. Whereas 1-bromo-2-iodobenzene afforded almost identical results (entry 3), 1-chloro-2-iodobenzene gave a much lower yield at 25% (entry 4), which suggested that the second intramolecular amination was less efficient with chloride than iodide and bromide. In the cases of both 1,2-dibromobenzene (entry 5) and 1-bromo-2-chlorobenzene (entry 6), incomplete reactions were observed. Not surprisingly, 1,2-dichlorobenzene (entry 7) failed to produce any desired product. A number of commercially available 1,2-dihaloarenes with different substitutions were then investigated (entries 8−11). In all cases, the desired benzimidazoles **8**−**11** were isolated in 15−53% yield. It is noteworthy that even a heterocylic aryl-dihalide afforded the desired benzimidazole **12** in low yield. The reactivity of the dihaloarenes

TABLE 3. Benzimidazole Synthesis with Substituted Guanidines



entry	R	conditions	product	yield[a]
1		130 °C, 16h	**13**	59%
2		150 °C, 16h	**14**	47%
3		165 °C, 16h	**15**	36%
4		165 °C, 16h	**16**	29%
5		165 °C, 16h	**17**	32%

[a] Isolated yield.

appears in the order of 1,2-di-I≈1-Br-2-I > 1,2-di-Br > 1-Cl-2-I > 1-Br-2-Cl.

The reaction scope with respect to the susbstituted guanidine was next explored. Either *N*,*N*-bis-substituted (Table 3, entries 1 and 2) or *N*-monosubstituted (entries 3−5) guanidines could be utilized to prepare 1-*H*-2-amino-benzimidazoles. It appears that guanidines with electron-withdrawing substituents require higher temperature for the reaction to proceed (entries 4 and 5).

We were pleased to find that this methodology was also applicable to the preparation of 2-alkyl (Table 4, entries 1 and 2), aryl (entries 3−5), or heteroaryl (entries 6 and 7) benzimidazoles. In these cases, higher temperature (170 °C) with NMP as the solvent was usually necessary to drive the reactions to completion. In addition to the desired benzimidazole products, the uncyclized protodehalogenated amidines (**18b**−**23b**) were the main side products. Nevertheless, this method provided useful yields of the desired benzimidazoles in one step. Substitution at the 1-*N* position was tolerated to afford a good yield of the *N*-substituted benzimidazole **25** (entry 8), which was often hard to prepare.[17b] Particularly interesting was entry 9, where tricyclic annulated benzimidazole **26** was prepared in one step, which required a much longer sequence using alternative methods.[23]

In conclusion, we have demonstrated that CuI/**L5** (*N*,*N*′-dimethylethylenediamine) is an efficient catalyst system for the guanidinylation of aryliodides. Using this catalyst system, a number of 1-*H*-2-amino-benzimidazoles were prepared from readily available 1,2-dihaloarenes and guanidines in one step in modest yields. The methodology is also suitable for the synthesis of other 2-substituted benzimidazoles.

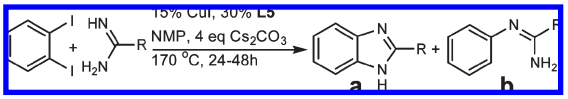
Experimental Section

General Procedure for the Monoamination (1)[21b]**.** A Schlenk tube equipped with a strong magnetic stirring bar was charged with CuI (38 mg, 0.2 mmol, 0.1 equiv), morpholine-4-carboxamidine hydrobromide (588 mg, 2.8 mmol, 1.4 equiv), 3,4-dichloro-iodobenzene (545 mg, 2 mmol, 1.0 equiv), and

(22) Compound **7** and its analogs are exceptionally polar and only dissolve in high polarity solvents such as MeOH, DMF, and DMSO. Reverse-phase preparatory HPLC was utilized for purification and might have contributed to the lower isolated yield.

(23) (a) McClure, J. R.; Custer, J. H.; Schwarz, H. D.; Lill, D. A. *Synlett* **2000**, *5*, 710–712. (b) Tan, K. L.; Vasudevan, A.; Bergman, R. G.; Ellman, J. A.; Souers, A. J. *Org. Lett.* **2003**, *5*, 2131–2134.

TABLE 4. Benzimidazole Synthesis with 2-Alkyl or Aryl Amidines



entry	R	products	yield[a] (a, b)
1	Pr^i	**18a +18b**	66%, 20%
2	Bu^t	**19a + 19b**	64%, 23%
3	Ph	**20a + 20b**	45%, 31%
4	O_2N	**21a + 21b**	30%, 15%
5	O, O	**22a + 22b**	41%, 15%
6	O	**23a + 23b**	23%, 35%
7	N	**24a**	32%, -[b]
8	NH, Ph, HN–Ph	**25**	58%, -[c]
9	NH, NH	**26**	52%, -[c]

[a] Isolated yield. [b] The side product decomposed upon isolation. [c] Not observed.

Cs_2CO_3 (1.95 g, 6 mmol, 3 equiv). The Schlenk tube was evacuated and backfilled with N_2 three times. Under N_2 atmosphere, *N*,*N*′-dimethylethylenediamine (35 mg, 0.4 mmol, 0.2 equiv) and 1,4-dioxane (5 mL) were added via syringe. The tube was sealed and stirred at 100 °C for 24 h and then cooled to room temperature. The inorganic salt was filtered off and washed with EtOAc. The filtrate EtOAc solution was washed with water, dried over $MgSO_4$, and concentrated to afford the title compound as a tan solid (470 mg, 1.7 mmol, 86%). No further purification was performed. HPLC t_R: 1.97 min. ^{1}H NMR (500 MHz): δ 7.32 (d, J = 8.5, 1H), 6.99 (d, J = 2.1, 1H), 6.73 (dd, J = 8.5, 2.2, 1H), 3.79−3.72 (m, 4H), 3.50−3.30 (m, 4H). ^{13}C NMR (126 MHz, $CDCl_3$): δ 152.5, 149.8, 132.7, 130.9, 125.2, 124.8, 122.8, 66.5, 45.8. HRMS-ESI (*m*/*z*): $[M + H]^+$ calcd for $C_{11}H_{13}Cl_2N_3O$ 274.0508, found 274.0510.

General Procedure for Benzimidazole Synthesis through Tandem Aminations (7). A Schlenk tube equipped with a strong magnetic stirring bar was charged with CuI (57 mg, 0.3 mmol, 0.15 equiv), morpholine-4-carboxamidine hydrobromide (588 mg, 2.8 mmol, 1.4 equiv), and Cs_2CO_3 (2.6 g, 8 mmol, 4 equiv). The Schlenk tube was evacuated and backfilled with N_2 three times. Under N_2 atmosphere, DMA or NMP (5 mL), 1,2-diiodobenzene (659 mg, 2 mmol, 1.0 equiv) and *N*,*N*′-dimethylethylenediamine (53 mg, 0.6 mmol, 0.3 equiv) were added sequentially via syringe. The reaction mixture was stirred in a preheated oil bath at 150 °C for 24 h and then cooled to room temperature. EtOAc (25 mL) was added, and the suspension was stirred for 30 min. The inorganic salt was filtered off and washed with EtOAc. The volatile EtOAc was removed under reduced pressure, and the remaining DMA or NMP solution was directly loaded on a preparatory HPLC for purification to afford the title compound as a white solid (0.21 g, 1.03 mmol, 52%). HPLC t_R: 1.59 min. ^{1}H NMR (500 MHz, DMSO-d_6): δ 13.12 (s, 1H), 7.44 (dd, J = 5.9, 3.2, 2H), 7.28 (dd, J = 5.9, 3.2, 2H), 3.85−3.77 (m, 4H), 3.66−3.61 (m, 4H). ^{13}C NMR (126 MHz, DMSO-d_6): δ 150.5, 130.0, 123.4, 111.4, 64.8, 46.0. HRMS-ESI (*m*/*z*): $[M+H]^+$ calcd for $C_{11}H_{13}N_3O$ 204.1131, found 204.1134.

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Supporting Information Available: Experimental details and characterization of compounds **1**−**26**. This material is available free of charge via the Internet at http://pubs.acs.org.