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Microwave Enabled Umpulong Mechanism Based Rapid and Efficient Four- and Six-Component Domino Formations of 2-(2′-Azaaryl)imidazoles and *anti*-1,2-Diarylethylbenzamides

Bo Jiang,[†,‡] Xiang Wang,[†] Feng Shi,[†] Shu-Jiang Tu,*[,†,‡] Teng Ai,[§] Austin Ballew,[§] and Guigen Li*[,§]

[†]*School of Chemistry and Chemical Engineering, Xuzhou Normal University, Xuzhou, 221116, Jiangsu, P. R. China,* [‡]*College of Chemistry, Chemical Engineering, and Materials Science, Suzhou University, Suzhou, 215123, Jiangsu, P. R. China, and* [§]*Department of Chemistry and Biochemistry, Texas Tech University, Lubbock, Texas 79409-1061*

laotu@xznu.edu.cn; guigen.li@ttu.edu

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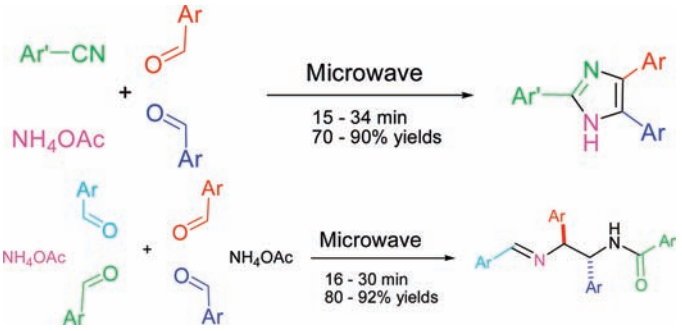

Concise and efficient six-component and four-component domino approaches to *anti*-1,2-diarylethylbenzamides and highly substituted 2-(2′-azaaryl)imidazoles have been developed under solvent-free and microwave-irradiation conditions. The reactions showed a broad scope of substrates in which a wide range of common commercial aromatic aldehydes and heteroaryl nitriles can be used. The syntheses were finished within short periods (15−34 min) with good to excellent chemical yields and stereoselectivity that avoided tedious workup isolations. New mechanisms involving an umpolung have been proposed for these two reaction processes.

Introduction

The development of efficient approaches to chemically and biologically important products from readily available inexpensive starting materials has been an active topic in modern organic chemistry.[1] In this regard, multicomponent domino reactions serving for the total synthesis of natural products and challenging building blocks are highly desired.[2] These reactions can avoid time-consuming and energy-costing processes that involve multistep syntheses, protection−deprotection, tedious workup, and purifications. Therefore, domino reactions are environmentally friendly and often proceed with excellent stereo- and chemoselectivities.[2,3]

In the past several years, we have been engaging in the development of new multicomponent domino reactions that

(1) (a) Tietze, L. F.; Hautner, F. In *Stimulating Concepts in Chemistry*; Vogtle, F., Stoddart, J. F., Shibasaki, M., Eds.; Wiley-VCH: Weinheim, 2000; pp 39−64. (b) Nicolaou, K. C.; Yue, E. W.; Oshima, T. in *The New Chemistry*; Hall, N., Ed.; Cambridge University Press: Cambridge, 2001; p186. (c) Tietze, L. F.; Brasche, G.; Gericke, K. *Domino Reactions in Organic Synthesis*; Wiley-VCH: Weinheim, 2006. (d) *New Avenues to Efficient Chemical Synthesis. Emerging Technologies*; Seeberger, P. H., Blume, T., Eds.; Springer: Heidelberg, 2007; Ernst Schering Foundation Symposium Proceedings 2006−3. (e) Tietze, L. F. *Chem. Rev.* **1996**, *96*, 115–136.

(2) (a) Snyder, S. A.; Breazzano, S. P.; Ross, A. G.; Lin, V; Zografos, S. L. *J. Am. Chem. Soc.* **2009**, *131*, 1753–1765. (b) Nicolaou, K. C.; Edmonds, D. J.; Bulger, P. G. *Angew. Chem., Int. Ed.* **2006**, *45*, 7134–7186. (c) Diguez-Vzquez, A.; Tzschucke, C. C.; Lam, W. Y.; Le, S. V. *Angew. Chem., Int. Ed.* **2008**, *47*, 209–212. (d) Muller, F. L.; Constantieux, T.; Rodriguez, J. *J. Am. Chem. Soc.* **2005**, *127*, 17176–17177.

(3) (a) Barluenga, J.; Jimenez-Aquino, A.; Valdes, C.; Aznar, F. *Angew. Chem., Int. Ed.* **2007**, *46*, 1529–1532. (b) Alex, K.; Tillack, A.; Schwarz, N.; Beller, M. *Angew. Chem., Int. Ed.* **2008**, *47*, 2304–2307. (c) Xia, M.; Xiang, G.-F.; Wu, B.; Han, Y.-F. *Adv. Synth. Catal.* **2008**, *350*, 817–821. (d) Waldmann, H.; Kuhn, M.; Liu, W.; Kumar, K. *Chem. Commun.* **2008**, 1211–1213.

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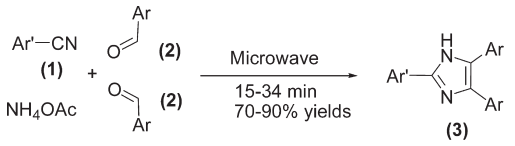
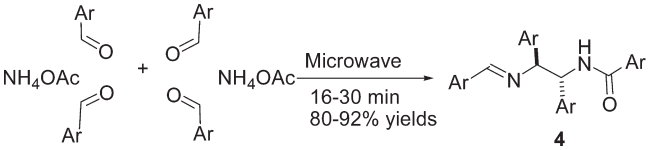
SCHEME 1. Four-Component Domino Synthesis of 2-(2′-Azaaryl)-imidazoles[a]



[a]Ar: **1a**, 2-pyridinyl; **1b**, 5-bromopyridine-2-yl; **1c**, 3-methylpyridine-2-yl; **1d**, 2-pyrazinyl; **1e**, 2-pyrimidinyl.

SCHEME 2. Six-Component Domino Synthesis of *anti*-Stilbenediamines



can provide easy access to new core structures of chemical and pharmaceutical interest.[4,5] During our study of this topic, we found an efficient microwave-accelerated synthesis of trisubstituted imidazoles (Scheme 1).[6] These products can serve not only as synthetic building blocks but also for supermolecular research on π-stacking and H-bonding formations.[7] In addition, the ligands containing 2-(2′-azaaryl)-imidazoles have also attracted widespread interest due to their ability to form complexes with transition metals.[8] In this paper, we would like to report a rapid four-component domino formation of 2-(2′-azaaryl)imidazoles and six-component domino formation of *anti*-1,2-diarylethylbenzamides[9] under microwave irradiation without the use of any organic solvents (Schemes 1 and 2).

It has been known that the synthesis of highly functionalized 2-(2′-azaaryl)imidazoles can be achieved via Negishi-type cross-coupling of 2-iodopyridine with imidazol-4-ylzinc reagent,[8a] three-component cyclocondensation of 1,2-diketones with picolinaldehydes in the presence of ammonium acetate,[8b] and the $NaBH_4$-catalyzed cyclization of 2-cyanopyridines.[8c] However, some limitations exist in these known methods. For example, Seto's method[8b] for formation of 2-(pyridin-2-yl)imidazoles (Pyim) is limited to the use of 1,2-diketones as starting materials that require multistep preparations involving benzoic condensation and oxidation reactions. We thus planned an alternative approach to these important compounds under more concise and efficient conditions.

TABLE 1. Optimization of Reaction Conditions

entry	ammonium salt	solvent	equiv	yield (%)
1	ammonium carbonate	solvent-free	3.0	59
2	ammonium chloride	solvent-free	3.0	38
3	ammonium formate	solvent-free	3.0	47
4	ammonium acetate	solvent-free	3.0	88

Results and Discussion

We started this synthesis by conducting the reaction of pyridine-2-carbonitrile **1a** with 4-chlorobenzaldehyde **2a** as the model case for condition optimization. As shown in Table 1, the use of ammonium acetate allowed the direct conversion of pyridine-2-carbonitrile **1a** into the corresponding 2-(2′-pyridinyl)imidazole **3a** in a yield of 88% under solvent-free and microwave-irradiation condition (Table 1, entry 4). Other ammonium salts gave much lower yields of 38−59% (Table 1, entries 1−3). The use of more than 3 equiv of ammonium acetate did not enhance chemical yields. We next studied solvent effects for this synthesis and found that DMF, benzene, acetic acid, and water resulted in moderate to good yields under MW irradiation for 20 min. However, higher yields were obtained when the reaction was performed under solvent-free condition.

We then investigated the substrate scope of this synthesis by subjecting the series of aromatic aldehyde **2b**−**k** to the reactions with pyridine-2-carbonitrile **1a** under the optimal condition. As shown in Table 2, the reaction of thiophene-2-carbaldehyde with pyridine-2-carbonitrile **1a** was complete within 20 min to give thienyl-substituted 2-(2′-azaaryl)-imidazoles **3l** in 72% yield. Similarly, 2-(2′-zaaryl)imidazoles **3b**−**k** were formed within 20−32 min in good to excellent yields of 74−90% (Table 2, entries 2−11). The similar situation exists for heteroaryl nitriles **1b**−**e** (5-bromopyridine-2-carbonitrile, **1b**; 3-methylpyridine-2-carbonitrile, **1c**; pyrazine-2-carbonitrile, **1d**; and pyrimidine-2-carbonitrile, **1e**) in which the reactions occurred rapidly to give the desired products **3r**−**v** in 76−84% yields (Table 2, entries 18−22). Among the products shown in Table 2, 2-(pyridin-2-yl)-imidazoles (entries 10, 11, 14, 22, and 25) are indeed very difficult to obtain via the known methods because 4-dimethylaminobenzaldehyde and 2-hydroxybenzaldehyde cannot serve as guest substrates to form symmetrical benzoins.

All starting materials of pyridine-2-carbonitriles and aryl aldehydes **2** employed for this synthesis are common inexpensive commercial chemicals, which avoids the use of 1,2-diketones and α-hydroxyketones for this synthesis. Our next plan is to choose special heteroaryl nitriles and aldehydes as the substrates to make lophine peroxides that are of particular interest to material sciences because of their wide range of chemiluminogenic activity[10] and to employ amino acid

(4) (a) Jiang, B.; Tu, S.-J.; Kaur, P.; Wever, W.; Li, G. *J. Am. Chem. Soc.* **2009**, *131*, 11660–11661. (b) Li, G.; Wei, H. X.; Kim, S. H.; Carducci, M. *Angew. Chem., Int. Ed.* **2001**, *40*, 4277–4280.

(5) (a) Tu, S.-J.; Jiang, B.; Jia, R. H.; Zhang, J. Y.; Zhang, Y.; Yao, C. S.; Shi, F. *Org. Biomol. Chem.* **2006**, *4*, 3664–3668. (b) Tu, S. J.; Jiang, B.; Zhang, Y.; Jia, R. H.; Zhang, J. Y.; Yao, C. S.; Shi, F. *Org. Biomol. Chem.* **2007**, *5*, 355–359. (c) Tu, S.-J.; Li, C. M.; Li, G. G.; Cao, L. J.; Shao, Q. Q.; Zhou, D. X.; Jiang, B.; Zhou, J. F.; Xia, M. *J. Comb. Chem.* **2007**, *9*, 1144–1148.

(6) (a) Lombardino, J. G.; Wiseman, E. H. *J. Med. Chem.* **1974**, *17*, 1182–1188. (b) Phillips, A. P.; White, H. L.; Rosen, S. *Eur. Pat. Appl. EP 58890*, **1982**; *Chem. Abstr.*, **1983**, *98*, 53894z.

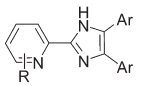
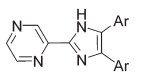
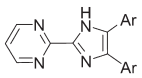
(7) (a) Wang, K.-Z.; Gao, L.-H.; Bai, G.-Y.; Jin, L.-P. *Inorg. Chem. Commun.* **2002**, *5*, 841–843. (b) Kimura, M.; Morioka, M.; Tsunenaga, M.; Hu, Z. Z. *New Technol. Med.* **2000**, *1*, 418–421. (c) Drew, M. G. B.; Tocher, D. A.; Chowdhury, K.; Chowdhury, S.; Datta, D. *New J. Chem.* **2004**, *28*, 323–325. (d) Pachhunga, K.; Therrien, B.; Kreisel, K. A.; Yap, G. P. A.; Kollipara, M. R. *Polyhedron* **2007**, *26*, 3638–3644.

(8) (a) Dobler, M. R. *Tetrahedron Lett.* **2003**, *44*, 7115–7117. (b) Seko, N.; Yoshino, K.; Yokota, K.; Tsukamoto, G. *Chem. Pharm. Bull.* **1991**, *39*, 651–657. (c) Yemada, M. K.; Kikugawa, Y. *Tetrahedron Lett.* **1969**, *36*, 3101–3104. (d) Chiswell, B.; Lions, F.; Morris, B. S. *Inorg. Chem.* **1964**, *3*, 110–114. (e) Kori, S.; Narisawa, S. *Asahi Garasu Kenkyu Hokoku* **1962**, *12*, 55–70.

(9) For the synthesis of *syn*-1,2-diarylethylbenzamides under different conditions and mechanism (longer reaction time and lower yields), see: Proskurnina, M. V.; Lozinskaya, N. A.; Tkachenko, S. E.; Zefirov, N. S. *Russ. J. Org. Chem.* **2002**, *38*, 1149–153.

(10) (a) White, E. H.; Harding, M. J. C. *J. Am. Chem. Soc.* **1964**, *86*, 5686–5687. (b) Kang, P.; Foote, C. S. *J. Am. Chem. Soc.* **2002**, *124*, 9629–9638.

TABLE 2. Domino Synthesis of 2-(2′-Azaaryl)imidazoles

Entry	3		Ar	Nitrile	Time /min	Yield[a] (%)
1	 **3a-3p**	**3a**	4-Chlorophenyl (**2a**)	**1a**	20	88
2		**3b**	4-Bromophenyl (**2b**)	**1a**	20	90
3		**3c**	4-Fluorophenyl (**2c**)	**1a**	20	84
4		**3d**	2-Chlorophenyl (**2d**)	**1a**	25	87
5		**3e**	Phenyl (**2e**)	**1a**	25	81
6		**3f**	4-Tolyl (**2f**)	**1a**	28	84
7	**3a-3l**; R = H	**3g**	4-Methoxyphenyl (**2g**)	**1a**	30	81
8	**3m-3o**; R = 5-Br	**3h**	Benzo[*d*][1,3]dioxol-5-yl (**2h**)	**1a**	26	84
9	**3p-3q**; R = 3-Me	**3i**	3,4,5-Trimethoxyphenyl (**2i**)	**1a**	32	74
10		**3j**	2-Hydroxyphenyl (**2j**)	**1a**	24	82
11		**3k**	4-Dimethylaminophenyl (**2k**)	**1a**	34	80
12		**3l**	2-Thienyl (**2l**)	**1a**	28	72
13		**3m**	Benzo[*d*][1,3]dioxol-5-yl (**2h**)	**1b**	26	81
14		**3n**	2-Hydroxyphenyl (**2j**)	**1b**	30	79
15		**3o**	2-Thienyl (**2l**)	**1b**	26	70
16		**3p**	3,4,5-Trimethoxyphenyl (**2c**)	**1c**	32	75
17		**3q**	2-Hydroxyphenyl (**2j**)	**1c**	30	78
18		**3r**	4-Chlorophenyl (**2a**)	**1d**	15	84
19	 **3r-3v**	**3s**	4- Fluorophenyl (**2c**)	**1d**	20	81
20		**3t**	4-Tolyl (**2f**)	**1d**	28	82
21		**3u**	Benzo[*d*][1,3]dioxol-5-yl (**2h**)	**1d**	32	76
22		**3v**	2-Hydroxyphenyl (**2j**)	**1d**	34	80
23	 **3w-3y**	**3w**	4-Methoxyphenyl (**2g**)	**1e**	28	82
24		**3x**	Benzo[*d*][1,3]dioxol-5-yl (**2h**)	**1e**	25	84
25		**3y**	2-Hydroxyphenyl (**2j**)	**1e**	26	83

[a]Isolated yields.

attached aromatic heteroaryl nitriles to generate unusual amino acids for peptide drug design and synthsis.[11]

After we achieved the above heteroaryl nitrile-based domino reaction, we then subjected benzonitrile to the reaction with aromatic aldehydes **2** under the similar conditions. Surprisingly, the unexpected products of *anti*-1,2-diarylethylbenzamides **4** were generated as a single stereoisomer in good to excellent yields of 81−92% (Scheme 2). Obviously, benzonitrile did not participate in the reaction with aromatic aldehydes.[9] The products have been unambigeously determined by X-ray structural analysis of single crystals of **3k** and **4i**, which were obtained by carefully evaporating solvents (see Supporting Information).

We then utilized various aromatic aldehydes to react with ammonium acetate under solvent-free and MW irradiation condition without the use of benzonitrile. As revealed in Table 3, 10 substituted *anti*-1,2-diarylethylbenzamides **4** were obtained in good to excellent yields (80−92%) within 16−30 min; these products are can be readily hydrolyzed by using sulfuric acid to give *anti*-stilbenediamines.[12−14]

On the basis of the above results, possible mechanisms have been proposed for the formations of trisubstituted 2-(2′-azaaryl)imidazole derivatives and 1,2-diarylethylbenzamides as shown in Schemes 3 and 4, respectively. The former involves the ring-closure cascade reactions that consist of initial condensation, nucleophilic addition, umpolung (**A** to **B**),[15] intramolecular nucleophilic addition (**B** to **C**), and

(11) (a) Schiller, P. W. *Prog. Med. Chem.* **1991**, *28*, 301–340. (b) Hruby, V. J.; Li, G.; Haskell-Luevano, C.; Shanderovich, M. *Pept. Sci.-Biopolym.* **1997**, *43*, 219–266.

(12) Corey, E. J.; Kuhnle, F. N. M. *Tetrahedron Lett.* **1997**, *38*, 8631–8634.

(13) (a) Kotti, S. R. S. S.; Timmons, C.; Li, G. *Chem. Biol. Drug Des.* **2006**, *67*, 101–114. (b) Lucet, D.; Gall, T. L.; Mioskowski, C. *Angew. Chem., Int. Ed.* **1998**, *37*, 2580–2627. (c) Samanta, S.; Liu, J.; Dodda, R.; Zhao, C.-G. *Org. Lett.* **2005**, *7*, 5321–5323. (d) Costa, A. M.; Jimeno, C.; Gavenonis, J.; Carroll, P. J.; Walsh, P. J. *J. Am. Chem. Soc.* **2002**, *124*, 6929–6941.

(14) (a) Bessonov, I. V.; Lozinskaya, N. A.; Katashova, V. R.; Proskurnina, M. V.; Zefirov, N. S. *Russ. Chem. Bull.* **2005**, *54*, 211. (b) Trippett, S. *J. Chem. Soc.* **1957**, 4407–4408.

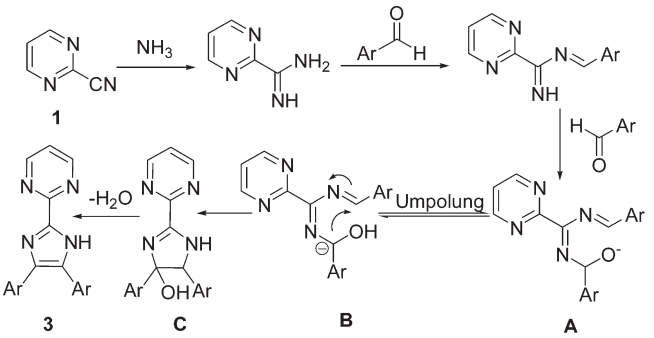
(15) (a) Enders, D.; Balensiefer, T. *Acc. Chem. Res.* **2004**, *37*, 534–541. (b) Enders, D.; Niemeier, O.; Henseler, A. *Chem. Rev.* **2007**, *107*, 5606–5655. (c) Johnson, J. S. *Angew. Chem., Int. Ed.* **2004**, *43*, 1326–1328. (d) Nair, V.; Bindu, S.; Sreekumar, V. *Angew. Chem., Int. Ed.* **2004**, *43*, 5130–5135. (e) Christmann, M. *Angew. Chem., Int. Ed.* **2005**, *44*, 2632–2634. (f) Zeitler, K. *Angew. Chem., Int. Ed.* **2005**, *44*, 7506–7510. (g) Marion, N.; Diez-Gonzalez, S.; Nolan, S. P. *Angew. Chem., Int. Ed.* **2007**, *46*, 2988–3000.

TABLE 3. Synthetic Results of 1,2-Diarylethylbenzamides

Entry	4	Ar	Time /min	Yield[a] (%)
1	4a	4-Chlorophenyl (**2a**)	16	89
2	4b	4-Bromophenyl (**2b**)	20	91
3	4c	4-Fluorophenyl (**2c**)	18	92
4	4d	2-Chlorophenyl (**2d**)	26	87
5	4e	Phenyl (**2e**)	30	88
6	4f	4-Tolyl (**2f**)	28	83
7	4g	4-Methoxyphenyl (**2g**)	30	85
8	4h	3,4,5-Trimethoxyphenyl (**2i**)	30	80
9	4i	3-Fluorophenyl (**2m**)	18	84
10	4j	2-Fluorophenyl (**2n**)	18	81

[a]Isolated yields.

SCHEME 3. Mechanism of the Formation of Trisubstituted 2-(2′-Azaaryl)imidazoles



dehydration (Scheme 3). The latter involves the condensation reaction to give imines, which is followed by subsequent intermolecular nucleophilic addition, umpolung (**D** to **E**), and nucleophilic reaction (**E** to **F**). The last step of latter mechanism involves 1,3-H transfer (**F** to **G**) leading to thermodynamically stable *anti*-diarylethylbenzamides **4** (Scheme 4).

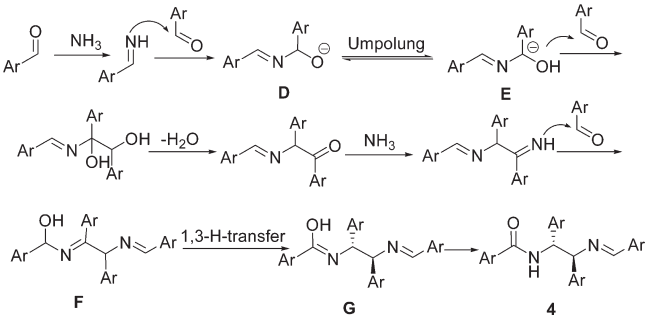
In summary, umpolung mechanism based concise and efficient six-component and four-component domino approaches to *anti*-1,2-diarylethylbenzamides and highly substituted 2-(2′-azaaryl)imidazoles were developed under solvent-free and microwave irradiation condition. The reactions showed a broad scope of substrates in which a wide range of common commercial aromatic aldehydes and heteroaryl nitriles can be used. The syntheses were complete within short periods (15−34 min) with good to excellent chemical yields and stereoselectivity that avoided tedious workup isolations. New mechanisms involving umpolung intermediates have been proposed for these two reaction processes.

Experimental Section

General. Microwave irradiation was carried out with microwave oven Emrys Creator from Personal Chemistry, Uppsala, Sweden.

Representative Example. 4,5-Bis(4-chlorophenyl)-1*H*-2-(2′-pyridyl)imidazole (3a). Microwave Heating. 2-Cyanopyridine (**1a**, 1.1 mmol, 0.111 g, 1.1 equiv) was introduced in a 10-mL Emrys reaction vial, and 4-chlorobenzaldehyde (**2a**, 2 mmol, 0.28 g, 2.0 equiv) and NH_4OAc (6 mmol, 0.46 g, 3.0 equiv) were

SCHEME 4. Mechanism of the Formation of *anti*-Diarylethylbenzamides



then successively added. Subsequently, the reaction vial was capped and then stirred for 20 s. The mixture was irradiated (initial power 100 W and maximum power 200 W) at 150 °C until TLC (petroleum ether/acetone, 4:1 v/v) revealed that conversion of the starting material **2a** was complete (20 min). The reaction mixture was then cooled to room temperature and diluted with cold water (40 mL). The solid product was collected by Büchner filtration and was purified by flash column chromatography (silica gel, mixtures of petroleum ether/ethyl acetate, 10:1 v/v) to afford the desired pure products **3a** as an pale green solid. Mp: 210−212 °C. IR (KBr, ν, cm^{-1}): 3056 (brs), 2959, 1594, 1565, 1505, 1401, 1293, 1244, 1130, 1092, 1012, 883, 823. ^1H NMR (DMSO-d_6, 400 MHz) (δ, ppm): 9.21 (s, 1H, NH), 8.45 (d, J = 7.2 Hz, 1H, Py-H), 8.03 (d, J = 8.4 Hz, 2H, Ar−H), 7.93 (d, J = 8.8 Hz, 2H, Ar−H), 7.86 (d, J = 8.8 Hz, 1H, Py-H), 7.64 (d, J = 8.8 Hz, 2H, Ar−H), 7.56 (d, J = 8.8 Hz, 2H, Ar−H), 7.03−7.00 (m, 1H, Py-H), 6.85−6.82 (m, 1H, Py-H). ^{13}C NMR (DMSO-d_6, 100 MHz) (δ, ppm): 151.3, 137.9, 135.7, 134.8, 133.4, 129.7, 129.5, 129.0, 128.8, 128.3, 127.0, 122.6, 121.3, 117.6, 114.7. HRMS (ESI), 366.0560: m/z calcd for $C_{20}H_{14}Cl_2N_3$, found 366.0559.

***N*-(*E*-2-(4-Chlorobenzylideneamino)-1,2-bis(4-chlorophenyl)-ethyl)-4-chlorobenzamide (4a).** 4-Chlorobenzaldehyde (**2a**, 4 mmol, 0.59 g, 4.0 equiv) was introduced in a 10-mL Emrys reaction vial, and NH_4OAc (8 mmol, 0.616 g, 4.0 equiv) was then added. Subsequently, the reaction vial was capped and then stirred for 20 s. The mixture was irradiated (initial power 100 W and maximum power 200 W) at 150 °C until TLC (petroleum ether/acetone, 4:1) revealed that conversion of the starting material **2a** was complete (16 min). The reaction mixture was then cooled to room temperature and then diluted with cold water (40 mL). The solid product was collected by Büchner filtration and was purified by recrystallization from 95% EtOH to afford the desired pure products **4a** as a white solid. Mp: 242−243 °C (lit. mp 249 °C).[14b] IR (KBr, ν, cm^{-1}): 3365, 3084, 1651, 1578, 1510, 1493, 1487, 1276, 1143, 1009, 971, 893. ^1H NMR (400 MHz, DMSO-d_6) (δ, ppm): 8.98(d, J = 9.2 Hz, 1H, NH), 8.07 (s, 1H, CH), 7.67−7.31 (m, 16H, ArH), 5.55 (t, J = 9.2 Hz, 1H, CH), 4.80(d, J = 9.6 Hz, 1H, CH).

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Supporting Information Available: ^1H and ^{13}C NMR spectra of all pure products. This material is available free of charge via the Internet at http://pubs.acs.org.