

One-Pot Synthesis of Pyrimido[1,6-*a*]indol-1(2*H*)-one Derivatives by a Nucleophilic Addition/Cu-Catalyzed N-Arylation/Pd-Catalyzed C–H Activation Sequential Process

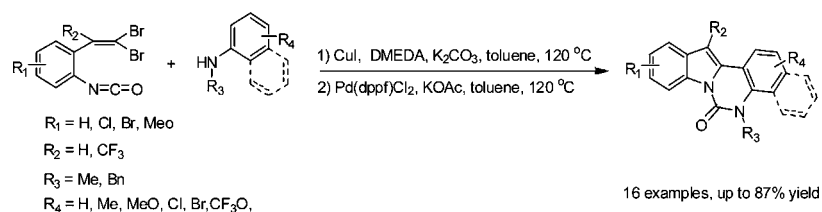
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Received May 6, 2010

ABSTRACT



A novel and convenient one-pot synthesis of pyrimido[1,6-*a*]indol-1(2*H*)-one derivatives through a nucleophilic addition/Cu-catalyzed N-arylation/Pd-catalyzed C–H activation sequential process is described. The reaction of easily prepared *ortho-gem*-dibromovinyl isocyanates with *N*-alkylanilines gave the desired indole derivatives in moderate to good yields.

The indole framework represents a privileged structural motif of important value in biologically active natural products and pharmaceutical compounds.¹ The indole-incorporated pyrimido[1,6-*a*]indol-1(2*H*)-one derivatives could be applied as fluorescent materials,² 5-HT₃ receptor antagonists,³ topoisomerase II inhibitors,⁴ etc. (Figure 1, A–C). However, the methods for the assembly of these molecules were limited.^{2–5} Therefore,

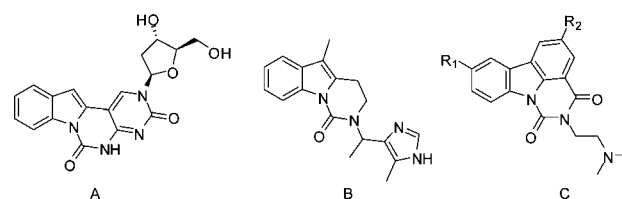


Figure 1. Several pyrimido[1,6-*a*]indol-1(2*H*)-one derivatives reported as biologically active compounds and pharmaceutical products.

more efficient and facile routes, to synthesize these useful molecules under mild conditions, are needed.

Cascade, tandem, and domino reactions could offer the opportunity to access final products with high efficiency from

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simple starting materials because they avoid the tedious step by step separations and purifications of intermediates and reduce the amount of pollutant waste, compared to traditional stepwise synthesis. For these reasons, most of the recently reported methods for the synthesis of indoles were based on the use of these types of reactions.⁶

In the past decade, Cu-mediated sp² C–X (X = N, O, S, etc.) bond formation reactions have drawn considerable attention for their efficiency and low cost.⁷ Recently, these Cu-catalyzed reactions have been successfully applied to the assembly of various heterocyclic compounds via one-pot strategies.⁸ Our research group has also reported alternative protocols for the synthesis of heterocycles based on Cu-catalyzed coupling reactions.⁹ Aza-accumulated olefins such as carbodiimides^{9c,g} and isothiocyanates,^{8a,9a,d} which could easily undergo nucleophilic addition, were employed as common substrates in Cu-catalyzed one-pot protocols. However, the similar protocols using isocyanates as nucleophilic acceptors were not well documented.

The C–H activation approach, for its sustainable and environmentally benign features, has received substantial attention.¹⁰ Most notably, direct arylation on the *ortho* positions of anilines via C–H activation have become the focus of many research groups.¹¹ However, there were only several reports of reactions in which direct arylations were involved with another coupling process.^{12,14h}

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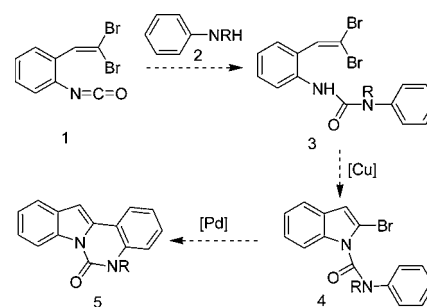
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More recently, *ortho-gem*-dihalovinylanilines¹³ have been developed to synthesize 2-substituted indole derivatives via domino processes.^{14–16} 2-Substituted benzofuran^{13,14f,17b} and benzothiophenes¹⁷ were also obtained from the corresponding *ortho-gem*-dihalovinyl phenols and thiophenols. However, to the best of our knowledge, the substituents on the *ortho* position of the *gem*-dihalostyrene were all nucleophilic groups, and none of the reactions of *gem*-dihalostyrene with electrophilic ones on the *ortho* position were reported.

Accordingly, we envisioned an addition/N-arylation/C–H activation sequential process to access pyrimido[1,6-*a*]indol-1(2*H*)-one derivatives in one pot, from easily prepared *ortho-gem*-dibromovinylisocyanates and *N*-alkyl-anilines. Herein, we would like to report the results (Scheme 1).

Scheme 1. Strategy for the Synthesis of Pyrimido[1,6-*a*]indol-1(2*H*)-one Derivatives



The proposed sequential process was first examined using *ortho-gem*-dibromovinylisocyanate **1a** and *N*-methyl aniline **2a** to form the addition product **3a** nearly quantitatively.¹⁸ As we expected, the *N*-arylation of **3a** catalyzed by CuI occurred to

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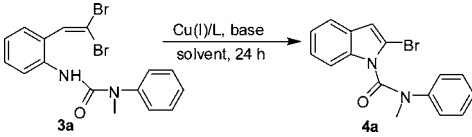
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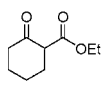
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provide corresponding 2-bromoindole **4a**, and the intramolecular direct arylation of **4a** catalyzed by PdCl₂ was successful to access the desired product **5a**. Then we searched for the optimized cyclization conditions of C–N bond formation using **3a** as a model substrate, as shown in Table 1.

Table 1. Optimization of the C–N Bond Formation Reaction Conditions^a



entry	Cu(I)	ligand	base	temp (°C)	solvent	yield (%)
1	CuI	1,10-Phen ^b	K ₂ CO ₃	120	Toluene	83
2	CuI	L-Proline	K ₂ CO ₃	120	Toluene	75
3	CuI		K ₂ CO ₃	120	Toluene	62
4	CuI	DMEDA ^c	K ₂ CO ₃	120	Toluene	92
5	CuI	No ligand	K ₂ CO ₃	120	Toluene	53
6	CuI	DMEDA	CS ₂ CO ₃	120	Toluene	49
7	CuI	DMEDA	KOAc	120	Toluene	35
8	CuI	DMEDA	Et ₃ N	120	Toluene	n.r. ^d
9	CuI	DMEDA	K ₂ CO ₃	80	Toluene	47
10	CuI	DMEDA	K ₂ CO ₃	100	Toluene	69
11	CuI	DMEDA	K ₂ CO ₃	110	Dioxane	86
12	CuI	DMEDA	K ₂ CO ₃	90	CH ₃ CN	23
13	CuI	DMEDA	K ₂ CO ₃	120	DMF	17
14	CuBr	DMEDA	K ₂ CO ₃	120	Toluene	75
15	Cu ₂ O	DMEDA	K ₂ CO ₃	120	Toluene	27

^a Reaction conditions: 3-(2-(2,2'-dibromovinyl)phenyl)-1-methyl-1-phenylurea **3a** (0.5 mmol), Cu source (0.05 mmol), ligand (0.1 mmol), and base (1.0 mmol) in solvent (4.0 mL) under N₂, for 24 h. ^b 1,10-Phen = 1,10-phenanthroline. ^c DMEDA = *N,N'*-dimethylethylenediamine. ^d n.r. = no reaction.

Initial studies focused on the selection of an efficient ligand. After screening a range of common ligands, DMEDA was found to be superior (Table 1, entry 4). Then various bases and solvents were tested, and K₂CO₃ and toluene proved to be the most efficient. Reducing the temperature caused a diminution in the yield (entries 9 and 10). When the copper source was switched to CuBr or Cu₂O (entries 14 and 15), the yield decreased. So, the conditions described in entry 4 were selected as optimal.

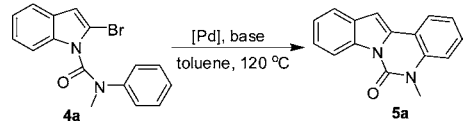
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To achieve the “one-pot” protocol, toluene was employed as the solvent for the direct arylation of **4a**. Different Pd sources were tested, and Pd(dppf)Cl₂ (Table 2, entry 2) was

Table 2. Optimization of the Second Cyclization: Intramolecular Direct Arylation Conditions^a



entry	Pd catalyst	base	yield (%)
1	PdCl ₂	K ₂ CO ₃	62
2	Pd(dppf)Cl ₂	K ₂ CO ₃	81
3	Pd(OAc) ₂	K ₂ CO ₃	25
4	Pd(dba) ₂	K ₂ CO ₃	trace
5	Pd(dppf)Cl ₂	CS ₂ CO ₃	65
6	Pd(dppf)Cl ₂	K ₃ PO ₄	52
7	Pd(dppf)Cl ₂	KOAc	90

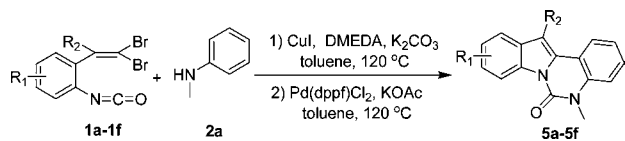
^a Reaction conditions: *N*-methyl-*N*-phenyl-2-bromo-1*H*-indole-1-carboxamide **4a** (0.5 mmol), Pd source (0.05 mmol), and base (1.5 mmol, 3.0 equiv) in toluene (4.0 mL) under N₂, for 24 h.

found to be the best catalyst for this second cyclization. Pd(OAc)₂ gave a much lower yield (entry 3), while Pd(dba)₂ failed to give any product (entry 4). KOAc was found to be the best base to promote the intramolecular direct arylation (entry 7). Then, the one-pot protocol was examined. When the formation of **4a** completed, Pd(dppf)Cl₂ and KOAc were directly added to the reaction mixture without further purification, to obtain **5a** successfully in good yield. However, using Pd(dppf)Cl₂ only without CuI could not provide **5a** from **3a** successfully. K₂CO₃ and KOAc also could not well promote the reaction from **3a** to **5a** independently.

Under the above optimized reaction conditions, various substituted *ortho-gem*-dibromovinyl isocyanates were used to test the generality of the reaction (Table 3). All the substituted *ortho-gem*-dibromovinyl isocyanates tested were converted smoothly to **5a–5f**. The method was proved to be general and efficient to prepare benzo-functionalized indole derivatives. Both electron-rich *ortho-gem*-dibromovinyl isocyanates (4,5-diMeO) (entry 2) and electron-deficient ones (5-Br, 4-Cl, 4-Br) could afford the corresponding products **5b–5e** (entries 3–5). It was worth mentioning that 3-substituted indoles could be provided from the corresponding isocyanate substrate in moderate yield (entry 6, 67%).

Then, various substituted *N*-alkyl-anilines were also investigated under the reaction conditions, as shown in Table 4. *N*-Methyl-anilines with electron-donating groups (4-Me, 4-MeO, 2-Me, 3,5-diMe, 3-Me) reacted efficiently with *ortho-gem*-dibromovinyl isocyanate **1a** and gave moderate to good yields (Table 4, entries 1–5). As we expected, **5k** and its isomer **5k'** were obtained as a mixture when *N*-methyl-3-Me aniline was used (3:2) (entry 5).¹⁹ The electron-withdrawing groups (4-Cl, 4-Br, 4-CF₃O) on the phenyl ring of *N*-methyl-aniline were also tolerated (entries 6–8). *N*-Benzyl-aniline worked even better than *N*-methyl-

Table 3. Nucleophilic Addition/Cu-Catalyzed N-Arylation/Pd-Catalyzed C–H Activation Sequential Reaction of Substituted *ortho-gem*-Dibromovinyl Isocyanates with *N*-Methyl-aniline^a



entry	R ₁	R ₂	t ₁ (h) ^b	t ₂ (h) ^c	yield (%)
1	H	H	24	24	78 (5a)
2	4,5-diMeO	H	18	24	65 (5b)
3	5-Br	H	24	24	57 (5c)
4	4-Cl	H	24	36	63 (5d)
5	4-Br	H	36	24	61 (5e)
6	H	CF ₃	24	24	67 (5f)

^a Reaction conditions: *ortho-gem*-dibromovinyl isocyanate **1** (0.5 mmol), *N*-methyl-aniline **2a** (0.55 mmol), CuI (0.05 mmol), DMEDA (0.1 mmol), and K₂CO₃ (1.0 mmol, 2.0 equiv) in toluene (4.0 mL), under N₂ at 120 °C for t₁, then Pd(dppf)Cl₂ (0.05 mmol) and KOAc (1.5 mmol), in toluene (4.0 mL), under N₂ at 120 °C for t₂. ^b Time for Cu-Catalyzed N-arylation. ^c Time for Pd-catalyzed C–H activation.

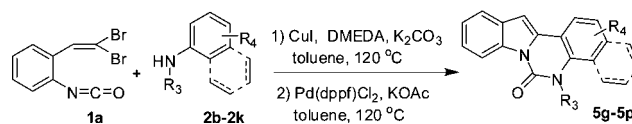
aniline (entry 9, 87%). *N*-Methyl-1-naphthylamine also participated in the sequential process leading to indole derivative **5p** in moderate yield (entry 10, 69%). As a limitation of our method, *N*-methyl-aniline, bearing a strongly electron-withdrawing group on the phenyl ring, such as 4-NO₂ and 4-Ac, which had a detrimental effect on nucleophilicity, failed to react with *ortho-gem*-dibromovinyl isocyanate to obtain the addition product. The addition of diphenylamine with *ortho-gem*-dibromovinyl isocyanate was also unsuccessful, due to its high steric hindrance.

In summary, we have developed a novel and convenient one-pot protocol to synthesize pyrimido[1,6-*a*]indol-1(2*H*)-one derivatives through a nucleophilic addition/Cu-catalyzed N-arylation/Pd-catalyzed C–H activation sequential process.

(18) We also examined unprotected anilines, such as aniline and 4-methyl-aniline. The corresponding urea was obtained, but the Cu-catalyzed N-arylation was unsuccessful.

(19) The regioisomer ratio was based on the ¹H NMR spectrum, and the configuration of the isomers was determined by NOE.

Table 4. Nucleophilic Addition/Cu-Catalyzed N-Arylation/Pd-catalyzed C–H Activation Sequential Reaction of *ortho-gem*-Dibromovinyl Isocyanate with Various Substituted *N*-Alkyl-anilines^a



entry	R ₃	R ₄	t ₁ (h) ^b	t ₂ (h) ^c	yield (%)
1	Me	4-Me	24	24	80 (5g)
2	Me	4-MeO	26	24	67 (5h)
3	Me	2-Me	30	24	62 (5i)
4	Me	3,5-diMe	26	36	64 (5j)
5	Me	3-Me	30	36	68 (5k , 5k')
6	Me	4-Cl	26	24	77 (5l)
7	Me	4-Br	24	24	71 (5m)
8	Me	4-CF ₃ O	30	36	69 (5n)
9	Bn	H	24	24	87 (5o)
10	Me	1-Naphthyl	16	24	69 (5p)

^a Reaction conditions: *ortho-gem*-dibromovinyl isocyanate **1a** (0.5 mmol), *N*-alkyl-aromatic amine **2** (0.55 mmol), CuI (0.05 mmol), DMEDA (0.1 mmol), and K₂CO₃ (1.0 mmol) in toluene (4.0 mL), under N₂ at 120 °C for t₁, then Pd(dppf)Cl₂ (0.05 mmol) and KOAc (1.5 mmol), in toluene (4.0 mL), under N₂ at 120 °C for t₂. ^b Time for Cu-Catalyzed N-arylation. ^c Time for Pd-catalyzed C–H Activation.

The reaction is applicable to a variety of *ortho-gem*-dibromovinyl isocyanates and *N*-alkyl-anilines, and moderate to good yields are attained. The products are potentially useful. Further investigations into synthetic applications are underway.

Acknowledgment. This work was financially supported by the Specialized Research Fund for the Doctoral Program of Higher Education of China (20060335036) and Zhejiang Provincial Natural Science Foundation of China under Grant No. Y406049.

Supporting Information Available: Experimental procedure, characterization data, and copies of ¹H NMR and ¹³C NMR spectra for all new compounds. This material is available free of charge via the Internet at <http://pubs.acs.org>.

OL101041E