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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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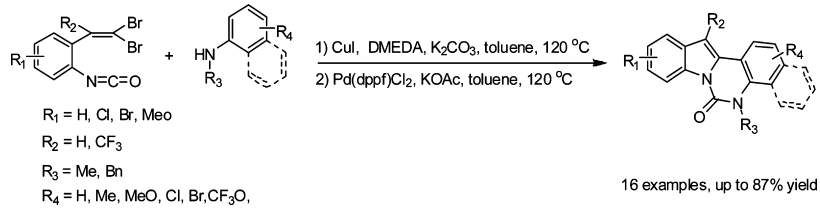
ABSTRACT



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-HT_3 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, more efficient and facile routes, to synthesize these useful molecules under mild conditions, are needed.

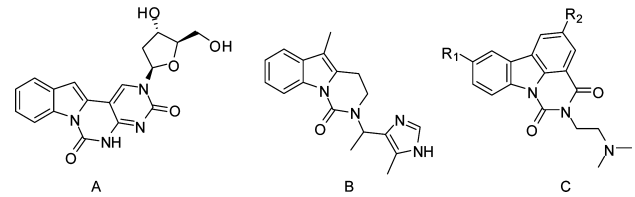


Figure 1. Several pyrimido[1,6-*a*]indol-1(2*H*)-one derivatives reported as biologically active compounds and pharmaceutical products.

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]

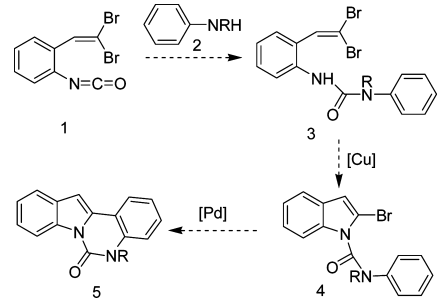
In the past decade, Cu-mediated sp^2 C−X (X = N, O, S, etc.) bond formation reactions have drawn considerable attention for their efficiency and low cost.[7] Recently, these Cu-catalyzed reactions have been successfully applied to the assembly of various heterocyclic compounds via one-pot strategies.[8] 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]

More recently, *ortho-gem*-dihalovinylanilines[13] have been developed to synthesize 2-substituted indole derivatives via domino processes.[14−16] 2-Substituted benzofuran[13,14f,17b] and benzothiophenes[17] 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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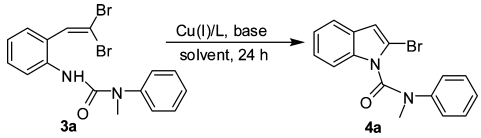
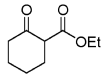
provide corresponding 2-bromoindole **4a**, and the intramolecular direct arylation of **4a** catalyzed by $PdCl_2$ was successful to access the desired product **5a**. Then we searched for the optimized cyclization conditions of C−N bond formation using **3a** as a model substrate, as shown in Table 1.

Table 1. Optimization of the C−N Bond Formation Reaction Conditions[a]



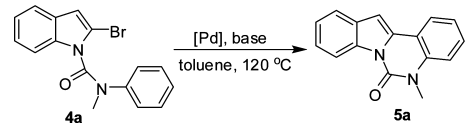
entry	Cu(I)	ligand	base	temp (°C)	solvent	yield (%)
1	CuI	1,10-Phen[b]	K_2CO_3	120	Toluene	83
2	CuI	L-Proline	K_2CO_3	120	Toluene	75
3	CuI		K_2CO_3	120	Toluene	62
4	CuI	DMEDA[c]	K_2CO_3	120	Toluene	92
5	CuI	No ligand	K_2CO_3	120	Toluene	53
6	CuI	DMEDA	Cs_2CO_3	120	Toluene	49
7	CuI	DMEDA	KOAc	120	Toluene	35
8	CuI	DMEDA	Et_3N	120	Toluene	n.r.[d]
9	CuI	DMEDA	K_2CO_3	80	Toluene	47
10	CuI	DMEDA	K_2CO_3	100	Toluene	69
11	CuI	DMEDA	K_2CO_3	110	Dioxane	86
12	CuI	DMEDA	K_2CO_3	90	CH_3CN	23
13	CuI	DMEDA	K_2CO_3	120	DMF	17
14	CuBr	DMEDA	K_2CO_3	120	Toluene	75
15	Cu_2O	DMEDA	K_2CO_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 N_2, 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 K_2CO_3 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 Cu_2O (entries 14 and 15), the yield decreased. So, the conditions described in entry 4 were selected as optimal.

To achieve the "one-pot" protocol, toluene was employed as the solvent for the direct arylation of **4a**. Different Pd sources were tested, and $Pd(dppf)Cl_2$ (Table 2, entry 2) was 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, $Pd(dppf)Cl_2$ and KOAc were directly added to the reaction mixture without further purification, to obtain **5a** successfully in good yield. However, using $Pd(dppf)Cl_2$ only without CuI could not provide **5a** from **3a** successfully. K_2CO_3 and KOAc also could not well promote the reaction from **3a** to **5a** independently.

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



entry	Pd catalyst	base	yield (%)
1	$PdCl_2$	K_2CO_3	62
2	$Pd(dppf)Cl_2$	K_2CO_3	81
3	$Pd(OAc)_2$	K_2CO_3	25
4	$Pd(dba)_2$	K_2CO_3	trace
5	$Pd(dppf)Cl_2$	Cs_2CO_3	65
6	$Pd(dppf)Cl_2$	K_3PO_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 N_2, for 24 h.

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-CF_3O) on the phenyl ring of *N*-methyl-aniline were also tolerated (entries 6−8). *N*-Benzyl-aniline worked even better than *N*-methyl-

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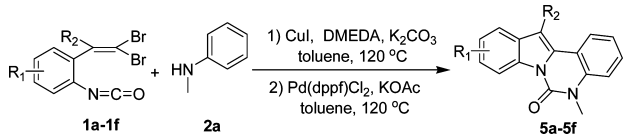
Table 3. Nucleophilic Addition/Cu-Catalyzed N-Arylation/Pd-Catalyzed C−H Activation Sequential Reaction of Substituted *ortho-gem*-Dibromovinyl Isocyanates with *N*-Methyl-aniline[a]



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 K_2CO_3 (1.0 mmol, 2.0 equiv) in toluene (4.0 mL), under N_2 at 120 °C for t_1, then Pd(dppf)Cl_2 (0.05 mmol) and KOAc (1.5 mmol), in toluene (4.0 mL), under N_2 at 120 °C for t_2. [b] Time for Cu-Catalyzed N-arylation. [c] Time for Pd-catalyzed C−H activation.

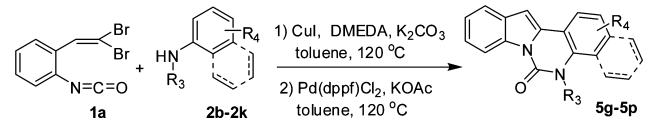
aniline (entry 9, 87%). *N*-Methyl-1-naphthylamine also participated in the sequential process leading to indole derivative **5p** in moderate yield (entry 10, 69%). As a limitation of our method, *N*-methyl-aniline, bearing a strongly electron-withdrawing group on the phenyl ring, such as 4-NO_2 and 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 ^{1}H 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_3O	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 K_2CO_3 (1.0 mmol) in toluene (4.0 mL), under N_2 at 120 °C for t_1, then Pd(dppf)Cl_2 (0.05 mmol) and KOAc (1.5 mmol), in toluene (4.0 mL), under N_2 at 120 °C for t_2. [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.

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

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