International Journal of Organic Chemistry, **2013, 3, 185-189**
http://dx.doi.org/10.4236/ijoc.2013.33023 Published Online September 2013 (http://www.scirp.org/journal/ijoc)

Salen-Cu(II) Complex Catalyzed *N*-Arylation of Imidazoles under Mild Conditions

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Received June 26, 2013; revised July 28, 2013; accepted August 8, 2013

ABSTRACT

Three inexpensive and air-/moisture-stable Salen-Cu complexes **1-3** were evaluated to be a novel class of catalysts for the *N*-arylation of imidazoles with aryl halides. A variety of aryl iodides, bromides underwent the coupling with imidazoles, promoted by the complex **3**, in moderate to excellent yields without the protection by an inert gas.

Keywords: Salen-Cu Complex; *N*-Arylation; Imidazole; Catalyze

1. Introduction

N-Aryl imidazoles and its derivatives are prevalent building blocks of numerous drugs, natural products and energetic materials, [1-5] and have been exploited as important precursors in *N*-heterocyclic carbene chemistry. [6,7] Therefore, their preparation has been attracted much attention. In recent years, the transition-metal palladium catalyzed *N*-arylation of imidazoles has made remarkable achievements, and shows relative mild reaction conditions, broad substrate scope and excellent functional-group tolerance [8-10]. However, in comparison with the use of costly palladium, it is desirable to develop more effective copper catalytic systems for *N*-arylation of imidazoles. The breakthroughs in this area, which were achieved by two research groups of Buchwald [11,12] and Taillefer, [13] respectively, have typically been driven by the implementation of new class of ligands and only catalytic amounts of copper metal under mild conditions. Following these pioneering works, several classes of mono-, bidentate, and polydentate chelators have thereby been developed to expedite the reaction rates and substantially lower the reaction temperature of Cu-based C-N coupling reaction [14-27]. In spite of the significant progress made in the aforementioned transformation, more efficient, air stable and cheaper ligands or metal-complexes for facilitating these coupling reactions under relatively milder conditions are still in demand.

Recently, our group had developed a series of effective catalysts, pyrrolecarbaldiminato-Cu complexes for C-N coupling reaction [28], and we previously reported Suzuki-Miyaura reaction catalyzed by Salen and half-salen palladium(II) complexes [29]. Although Salen-Pd complexes show low catalytic activity in the C-C coupling reaction, we reasonable assumed that Salen-Cu complexes might be a class of effective catalysts for the C-N coupling reaction. Herein, we wish to report Salen-Cu complexes as catalysts for the *N*-arylation of imidazoles and its derivatives. This system contains several advantages as follows: 1) the complexes were easily synthesized from cheap starting materials, and stable in air and moisture; 2) the reaction condition was relatively milder and did not require the protection by an inert atmosphere; 3) the complexes worked well for aryl iodides, and bromides with moderate to excellent yields.

2. Results and Discussion

Initially, the catalytic activity of the complexes **1-3** evaluated by using the C-N coupling of 4-iodotoluene with imidazole as a model reaction in the presence of NaOH at 120˚C for 12 h in DMSO (**Scheme 1**). As expected, the three Salen-Cu complexes all exhibited high catalytic activity for this process, and which gave the desired product in 92% - 94% isolated yields (**Table 1**, entries 1-3). The coupling reaction did not occur in the absence of any catalyst (**Table 1**, entry 4). Subsequently, we select the complex **3** as catalyst to further investigate the effects of the other reaction conditions on the

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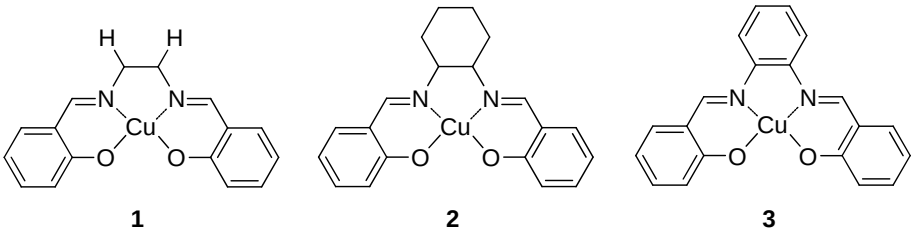
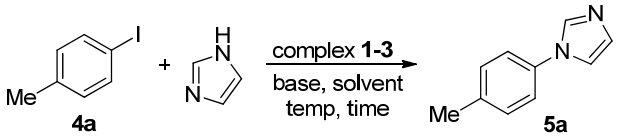


1 **2** **3**

Scheme 1. Structure of complexes 1 - 3.

Table 1. Optimization of the reaction conditions[a].



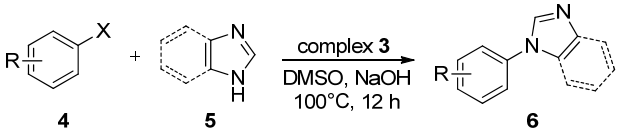
Entry	Complex	Base	Solvent	Temp (˚C)	Yield (%)[b]
1	-	NaOH	DMSO	120	0
2	1	NaOH	DMSO	120	92
3	2	NaOH	DMSO	120	94
4	3	NaOH	DMSO	120	94
5	3	NaOH	DMSO	100	94
6	3	NaOH	DMSO	80	88
7	3	Na_2CO_3	DMSO	100	0
8	3	K_3PO_4	DMSO	100	70
9	3	Cs_2CO_3	DMSO	100	95
10	3	NEt_3	DMSO	100	0
11	3	NaOH	DMF	100	43
12	3	NaOH	DMA	100	76
13	3	NaOH	H_2O	100	0
14	3	NaOH	DMSO	100	82[c]

[a] Reaction conditions: 4-ioidotoluene (0.5 mmol), imidazole (1.0 mmol), complex 1-3 (10 mol%), base (1.0 mmol), and solvent (1 ml), reaction time 12 h. [b] Isolated yields. [c] Complex **3** (5.0 mol%).

N-arylation reaction, including reaction temperature, base, solvent and catalyst loading. The results showed that 100˚C was enough for the coupling reaction to give 94% yield of the product (**Table 1**, entry 5), and the lower temperature decelerated the reaction rate. For example, 88% yield was obtained when the reaction was carried out at 80˚C (**Table 1**, entry 6). Base also plays an important role in the catalyst systems. Among various bases examined, K_3PO_4, and Cs_2CO_3 were all effective for the catalysis, and Cs_2CO_3 demonstrated the best improvement to give the corresponding product in 95% yield (**Table 1**, entries 8 and 9), but the use of Na_2CO_3 and organic base NEt_3 led to lower yields (**Table 1**, entries 7 and 10). However, NaOH was used in the following studies because it was less expensive than Cs_2CO_3. Solvent is another important factor affecting catalysis. It was found that DMSO performed as the prime solvent. Both DMF and DMA were not as good as DMSO. Meanwhile, H_2O was not suitable as a solvent (**Table 1**, entries 11-13). Furthermore, decreasing the loading of complex **3** from 10 mol% to 5 mol% led to a decrease of the yield (**Table 1**, entry 14). Finally, the combination of Salen-Cu complex **3** (10 mol%), NaOH (2 equiv.) at 100˚C for 12 h in DMSO was chosen as the optimal conditions for *N*-arylation of imidazole with 4-iodotoluene.

The scope of substrates was then investigated by using this catalytic system under the optimized reaction conditions. As shown in **Table 2**, In general, most of aryl iodides reacted with imidazole smoothly afforded the desired products in moderate to excellent yields. For example, 1-chloro-4-iodobenzene, 1-fluoro-4-iodobenzene and 4-iodo-1,1'-biphenyl led to the *N*-arylated products in 90% - 95% yields (**Table 2**, entries 6-8). When 1-(4-iodophenyl)ethanone and 1-iodo-4-nitrobenzene as coupling partners, the yields dropped to 72% and 60% respectively (**Table 2**, entries 4 and 5). Furthermore, the catalytic system could tolerate a variety of functional groups including the nitro, acetyl, and ether groups. Notably, sterically demanding *ortho* substituents such as 1-iodo-2-methylbenzene did not hamper the arylation reaction (**Table 2**, entry 3). Next, we were intrigued by the possibility of using aryl bromides as coupling partners. However, low yields were found under the previously optimized reaction conditions (**Table 2**, entries 9 and 10). In an endeavor to expand the scope of the methodology, this new catalytic system was applied to a variety of imidazole derivatives. To our delight, most of the aryl iodides reacted with the 1*H*-benzo[*d*]imidazole to provide the corresponding products in good to excellent yields (80% - 92%) under the optimized reaction conditions. Electron-withdrawing groups seemed to be a little beneficial for the catalysis compared to electron-donating ones. For example, 1-chloro-4-iodoben-zene, 1-fluoro-4-iodobenzene and 1-iodo-4-nitrobenzene afforded the corresponding arylated products in 80% - 92% yields (**Table 2**, entries 14, 16 and 17). Furthermore, aryl iodides with electron-donating could also be coupled with imidazole to give the products in good yields (**Table 2**, entries 11 and 15). Sterically hindered 1-iodo-2-methylbenzene afforded the product in low yield (**Table 2**, entry 13).

Table 2. *N*-Arylation of imidazole with aryl halides catalyzed by complex 3[a].



4 5 6

Entry	ArX R, X	HeT-NH	Product	Yield (%)[b]
1	H, I (**4b**)	imidazole (**5a**)	**6b**	98
2	4-OEt, I (**4c**)	**5a**	**6c**	83
3	2-Me, I (**4d**)	**5a**	**6d**	59
4	4-COMe, I (**4e**)	**5a**	**6e**	72
5	4-NO_2, I (**4f**)	**5a**	**6f**	60
6	4-Ph, I (**4g**)	**5a**	**6g**	95
7	4-Cl, I (**4h**)	**5a**	**6h**	90
8	4-F, I (**4i**)	**5a**	**6i**	98
9	4-Me, Br (**4j**)	**5a**	**6a**	19
10	4-NO_2, Br (**4k**)	**5a**	**6f**	49
11	Me, I (**4a**)	1H-benzo[d]imidazole (**5b**)	**6j**	87
12	H, I (**4b**)	**5b**	**6k**	83
13	2-Me, I (**4d**)	**5b**	**6l**	20[c]
14	4-NO_2, I (**4f**)	**5b**	**6m**	89
15	4-Ph, I (**4g**)	**5b**	**6n**	82
16	4-Cl, I (**4h**)	**5b**	**6o**	80
17	4-F, I (**4i**)	**5b**	**6p**	92
18	4-F, Br (**4l**)	**5b**	**6p**	21

[a] Reaction conditions: aryl halides (0.5 mmol), imidazoles (1.0 mmol), complex **3** (10 mol%), NaOH (1.0 mmol), and DMSO (1 ml), 100˚C, 12 h. [b] Isolated yields.

3. Conclusion

In summary, we have developed a novel and general catalytic method for *N*-arylation of imidazoles promoted by Salen-Cu(II) complex **3**. The system is efficient for the coupling of imidazoles and its derivatives with ArX (X = I, Br) to give moderation to excellent yields. The easy availability of the catalyst, mild reaction conditions, experimental simplicity, and broad substrate scope are the features of the catalytic method presented in the current paper. Further application of these Salen-Cu(II) complexes catalyzed organic reaction is currently ongoing in our laboratory.

4. Experimental

4.1. Materials and Instruments

All reactions were carried out under air using magnetic stirring unless otherwise noted. ^{1}H NMR spectral data were recorded on a Bruker DPX-400 spectrometer using TMS as internal standard and $CDCl_3$ as solvent. Mass spectra were recorded on GC-MS (Agilent 7890A/5975C) instrument under EI model. All other reagents were of analytical grade quality purchased commercially and used.

4.2. Synthesis of Complexes 1-3

$Cu(OAc)_2 \cdot H_2O$ (0.012 mol, 2.40 g) was added to a solution of substituted ethane-1,2-diamine (0.01 mol) and 2-hydroxybenzaldehyde (0.02 mol) in 35 ml methanol. The mixture was stirred at 60˚C for 5 h and then filtered. The precipitate was washed with dichloromethane. The solid product was collected and dried under vacuum to afford the desired complex **1-3**.

Complex **1** [30,32]: yield 65%. Anal. Calcd. for $C_{16}H_{14}CuN_2O_2$, %: C, 58.26; H, 4.28; N, 8.49; Found, %: C, 57.59; H, 4.38; N, 8.37.

Complex **2** [31,33,34]: yield 70%. Anal. Calcd. for $C_{20}H_{20}CuN_2O_2$, %: C, 62.57; H, 5.25; N, 7.30; Found, %: C, 62.72; H, 5.26; N, 7.49.

Complex **3** [34-37]: yield 72%. Anal. Calcd. for $C_{20}H_{14}CuN_2O_2$, %: C, 63.57; H, 3.73; N, 7.41; Found, %: C, 63.45; H, 3.81; N, 7.45.

4.3. General Procedure for *N*-Arylation of Imidazole with 4-Iodotoluene

To a 10 ml of sealed tube was added complex **3** (37.8 mg, 0.05 mmol), 4-iodotoluene (109 mg, 0.5 mmol), imidazole (68 mg, 1.0 mmol), NaOH (40 mg, 1.0 mmol), and DMSO (1 ml). The reaction mixture was reacted at 100˚C in a preheated oil bath for 12 h. The reaction mixture was cooled to r.t., diluted with 10 mL H_2O, and then the mixture was extracted with ethyl acetate (3 × 20 mL). The combined organic phases was washed with water and brine, dried over anhydrous Na_2SO_4, and concentrated in vacuo. The residue was purified by flash column chromatograph on silica gel (ethyl acetate/petroleum ether, 2:1 to pure ethyl acetate) to afford the target product (75 mg, 95% yield). 1-p-Tolyl-1H-imidazole (**6a**) [38-40], ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.81 (s, 1H), 7.27 (s, 4H), 7.24 (t, J = 1.2 Hz, 1H), 7.19 (s, 1H), 2.40 (s, 3H). GC-MS (EI): m/z = 158 $[M]^+$.

5. Acknowledgements

We gratefully acknowledge financial support of this work by the National Basic Research Program of China (973 Program: 2012CB722603), the National Natural Science Foundation of China (No. 21103114), the Ministry of Education Innovation Team (No. IRT1161), and Start-Up Foundation for Young Scientists of Shihezi

University (RCZX201012, RCZX201014, RCZX201015).

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