DOI: 10.1002/ejoc.200700281

Ultrasound-Promoted Reaction of 2-Chlorobenzoic Acids and Aliphatic Amines

Maite L. Docampo,[a] Rolando F. Pellón,*[a] Ana Estevez-Braun,[b] and Angel G. Ravelo[b]

Keywords: Ullmann condensation / Copper / Catalysis / Ultrasound / Aliphatic amines

An improvement to the use of DMF as a solvent for the condensation of 2-chlorobenzoic acids with aliphatic primary or secondary amines was described. A number of alkylaminobenzoic acids and dialkylaminobenzoic acids were synthesized in acceptable-to-good yield. The advantages of this procedure include readily available substrates, the use of an inexpensive copper powder without taking any precautions to exclude moisture under mild conditions and experimental ease. Furthermore, this condensation could also be achieved under nonclassical conditions by using ultrasonic irradiation at room temperature. We demonstrated that ultrasound-promoted condensation of 2-chlorobenzoic acid with aliphatic amines with the use of DMF as the solvent, especially in the case of secondary amines, affords products in high yields and reduces the reaction time to minutes. The results proved to be highly reproducible because the relevant sonochemical parameters were rigorously controlled.

Introduction

It is well-known that the substitution of a halogen atom onto a benzene ring does not take place easily. Classical Ullmann chemistry along with closely related methods have been known for a century and served well for C–N, C–S and C–O bond formation reactions.[1] These reactions require harsh prolonged heating at 200 °C or higher in the presence of stoichiometric amounts of simple Cu^{I} or Cu^{II} salts or oxides. The discovery of the Pd-catalyzed amination (Buchwald–Hartwig reaction)[2] reaction was a major breakthrough in the chemistry of amines, which opened access to huge numbers of previously inaccessible compounds. Nevertheless, high costs of palladium and some problematic functional group tolerances forced scientists to reconsider other transition-metal catalysts, mainly nickel and copper. A new interest in much cheaper and more practical copper-catalyzed chemistry was brought about by the observation that the appropriate ligand can modulate the reactivity of the catalyst and thus enable us to achieve more effective and more versatile catalytic systems.[3–12] The scope for CuI and amino acid catalyzed reaction of aryl halides with primary amines was explored in a recent publication.[13] The coupling reaction of electron-deficient aryl iodides with aliphatic primary amines occurred at 40 °C under the promotion of *N*-methylglycine. By using L-proline as a promoter, the coupling reaction of aryl iodides or -bromides with aliphatic primary amines proceeded at 60–90 °C. The steric hindrance of secondary amines makes the coupling reaction with aryl halides more sluggish than that of primary amines, and the coupling reaction between iodobenzene with morpholine or piperidine occurred at 90 °C. When less bulky pyrrolidine was used, the coupling reaction with aryl iodides proceeded at 65 °C.

In all cases the reaction conditions were CuI (0.5 mmol), amino acid (1 mmol), aryl halide (5 mmol), amine (7.5 mmol), K_2CO_3 (10 mmol for aryl iodide) or K_3PO_4 (10 mmol for aryl bromide), DMSO (3 mL) and *N*-methylglycine or L-proline, and the range of reaction time was 12 to 40 h. All reactions were carried out under a nitrogen atmosphere. DMF and DMSO were freshly distilled from CaH_2.

The combination of copper iodide and cesium acetate without ligands in the presence of DMSO or DMF as the solvent was used for the copper-mediated intermolecular amination of aryl iodides for the synthesis of secondary amines under an argon atmosphere.[14] Currently, the major restriction for this method is the low reactivity of the secondary acyclic amines, which is most likely due to steric reasons, so the scope of amines usually includes primary aliphatic and saturated secondary heterocycles, such as piperidine, piperazine, pyrrolidine, morpholine etc.

The majority of acids investigated were either iodobenzoic- or bromobenzoic acids, and only a few examples included the use of chlorobenzoic acids. Because chlorobenzoic acids are cheaper and more readily available than iodobenzoic- and bromobenzoic acids, we further explored the scope of the Ullmann condensation using chlorobenzoic

[a] Departamento de Química, Centro de Química Farmacéutica, Calle 21 y 200 Atabey, Playa. Apdo. 16042 CP 11600 Ciudad de la Habana, Cuba
Fax: +537-2736471
[b] Instituto Universitario de Bio-Orgánica Antonio González, Universidad de la Laguna,
Avda. Astrofisico F. Sanchez 2, 38206 La Laguna, Tenerife, España

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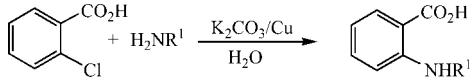
acids. Here we present the condensation of aliphatic primary or secondary amines with 2-chlorobenzoic acid derivatives in the presence of *N*,*N*-dimethylformamide as the solvent under ultrasonic irradiation conditions.

Results and Discussion

In a previous publication,[15] we analyzed the influence of some parameters on the condensation of 2-chlorobenzoic acid and substituted primary alkylamines in water as the solvent to yield the corresponding 2-(alkylamino)benzoic acids. It was observed that the reaction of 3 wt.-% of copper, 2.0 equiv. of alkylamine per mole of 2-chlorobenzoic acid and 1.0 equiv. of potassium carbonate afforded, after 5 h, the corresponding 2-(alkylamino)benzoic acid with yields superior to those reported in the literature with the use of other preparative procedures.[16–18] We also examined the effect of ultrasonic irradiation for the synthesis of these compounds, increasing the yields in a 20% rate after only 20 min[15] (Scheme 1).

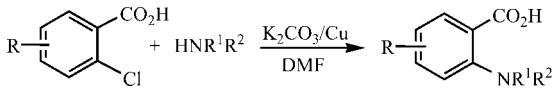


Scheme 1. Condensation of 2-cholorobenzoic acid with primary amines in water as the solvent.

We attempted the condensation reaction of secondary amines with 2-chlorobenzoic acid using ultrasound and water as the solvent, but condensation did not take place satisfactorily and salicylic acid was obtained in high yield.

It is assumed that copper ions can form chelates with 2-halobenzoic acids through the carboxyl group, thereby facilitating the nucleophilic substitution.[19] These copper chelates are destroyed in basic reaction media, and it is then not possible to obtain salicylic acid. We demonstrated that if pyridine is present there is a competition between the nucleophilic agent and the hydroxide group. So, we studied the effect of pyridine on the Ullmann condensation and found that pyridine acts as a ligand and reduces the reaction time.[20a,20b] These results suggest that pyridine stabilizes the intermediate copper complex.[20c] It may be possible that the secondary amines have the same effect as pyridine. After several trials, we found that this problem could be solved by using DMF as the solvent. DMF reduces the possibility that 2-chlorobenzoic acid can be hydrolyzed (salicylic acid formation).[20d]

As a first part of this work we studied the condensation of 2-chlorobenzoic acid derivatives with primary and secondary amines using DMF as the solvent (Scheme 2). To find the optimal reaction conditions, different reaction times and equivalents of K_2CO_3 were examined because these parameters play an important role in this reaction. We set out to test our proposal using 2-chlorobenzoic acid (1.0 equiv.) and propylamine (2.0 equiv., Table 1, Entry **3a**), 2-chloro-4-fluorobenzoic acid (1.0 equiv.) and morpholine (2.0 equiv., Table 1, Entry **8b**) or 2-chloro-4-nitrobenzoic acid (1.0 equiv.) and dipropylamine (2.0 equiv., Table 1, Entry **15b**) as the coupling partners with copper powder (3 wt.-%) as the catalyst.



Scheme 2. Condensation of 2-cholorobenzoic acids with aliphatic primary or secondary amines in DMF as the solvent.

Experimental results shown in Figures 1 and 2 demonstrate that better yields could be obtained with 1.0 equiv. of K_2CO_3 and a reaction time of 2 h in the case of **3a** (72%) and a 4 h reaction time in the cases of **8b** and **15b** (61 and 50%, respectively). It is noteworthy that when we used more than 1.0 equiv. of K_2CO_3 (basic pH) the yield of **3a** harshly diminished down to 12% with the use of 4.0 equiv. of K_2CO_3. Nevertheless, in the case of secondary amines (**8b** and **15b**) an excess of K_2CO_3 did not affect the reaction yield. This indicates that in the case of primary amines it is necessary that the reaction media has a neutral pH value.

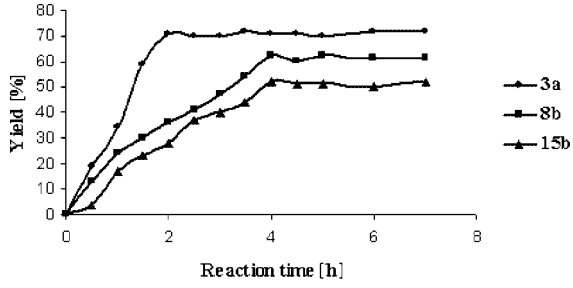


Figure 1. Plot of the yield [%] obtained in the condensation of 2-chlorobenzoic acid derivatives with a primary amine (**3a**), secondary cyclic amine (**8b**) or secondary acyclic amine (**15b**) as a function of reaction time [h].

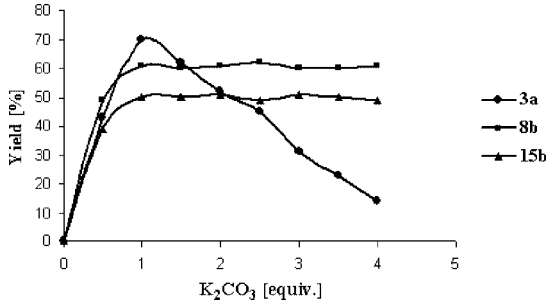
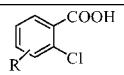
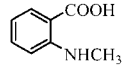
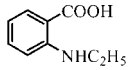
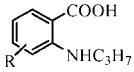
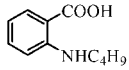
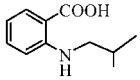
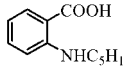
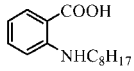
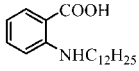
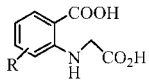
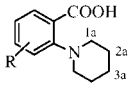
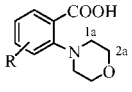
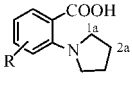
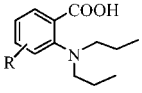
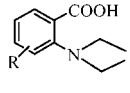


Figure 2. Plot of the yield [%] obtained in the condensation of 2-chlorobenzoic acids derivatives with a primary amine (**3a**), secondary cyclic amine (**8b**) or secondary acyclic amine (**15b**) as a function of K_2CO_3 equivalents.

On the basis of the above results, the condensation of different 2-chlorobenzoic acids with several primary and secondary aliphatic amines was tested, and the results are summarized in Table 1. For primary amines (Table 1, Entries **1a**–**16a**) it was found that glycine (amino acid) gave a better conversion relative to other alkylamines (compare Table 1, Entries **12a**–**16a**, and Entries **1a**–**11a**). Presumably, glycine acts as a ligand to form a copper complex, which

Table 1. Condensation of 2-chlorobenzoic acid derivatives (1.0 equiv.) with primary or secondary amines (2.0 equiv.) in DMF as the solvent with and without the use of ultrasound.

Entry		Amine	Yield[a] [%]	Products
1a	R = H	CH_3NH_2	73[b]/85[c]	
2a	R = H	$C_2H_5NH_2$	71[b]/83[c]	
3a	R = H		72[b]84[c]	
4a	R = 4-NO_2	$C_3H_7NH_2$	75[b]/86[c]	
5a	R = 5-NO_2		77[b]/89[c]	
6a	R = 4-F		74[b]/87[c]	
7a	R = H	$C_4H_9NH_2$	76[b]/88[c]	
8a	R = H	$iC_4H_9NH_2$	74[b]/86[c]	
9a	R = H	$C_5H_{11}NH_2$	75[b]/87[c]	
10a	R = H	$C_8H_{17}NH_2$	78[b]/89[c]	
11a	R = H	$C_{12}H_{25}NH_2$	80[b]/91[c]	
12a	R = H		85[b]/94[c]	
13a	R = 4-NO_2		86[b]/95[c]	
14a	R = 5-NO_2	$C_2H_3O_2NH_2$	88[b]/97[c]	
15a	R = 4-Cl		82[b]/91[c]	
16a	R = 5-Cl		87[b]/96[c]	
1b	R = 5-Cl		60[b]/86[c]	
2b	R = 5-NO_2		61[b]/90[c]	
3b	R = 4-NO_2		58[b]/83[c]	
4b	R = 4-F		57[b]/86[c]	
5b	R = 5-Cl		62[b]/83[c]	
6b	R = 5-NO_2		63[b]/89[c]	
7b	R = 4-NO_2		61[b]/85[c]	
8b	R = 4-F		61[b]/82[c]	
9b	R = 5-Cl		69[b]/91[c]	
10b	R = 5-NO_2		71[b]/92[c]	
11b	R = 4-NO_2		68[b]/88[c]	
12b	R = 4-F		66[b]/89[c]	
13b	R = 5-Cl		52[b]/78[c]	
14b	R = 5-NO_2		53[b]/79[c]	
15b	R = 4-NO_2		50[b]/72[c]	
16b	R = 4-F		51[b]/76[c]	
17b	R = 5-Cl		57[b]/81[c]	
18b	R = 5-NO_2		56[b]/82[c]	
19b	R = 4-NO_2		54[b]/75[c]	
20b	R = 4-F		54[b]/78[c]	

[a] Yield of isolated and recrystallized products from EtOH/H_2O. [b] Conventional conditions, reaction time: 2 h (**1a**–**16a**) and 4 h (**1b**–**20b**). [c] Ultrasonic irradiation (20 kHz, 350 W; 25 °C), reaction time: 25 (**1a**–**16a**), 15 (**1b**–**12b**) and 20 min (**13b**–**20b**).

facilitates the coupling reaction. In general, by using DMF the yields in the condensation of primary amines were significantly improved and the reaction time was reduced dramatically relative to those with water as the solvent.[15]

For secondary amines (Table 1, Entries **1b**–**20b**), we achieved moderate coupling yields (50–71%); this can be ascribed to the enhanced steric hindrance of the secondary amines relative to the primary amines. In the case of secondary acyclic amines (Table 1, Entries **13b**–**20b**), the yields obtained were higher than (51–57%) those reported for this coupling. Ma[13] described the coupling of iodobenzene with diethylamine, and the yield obtained was 21% in the presence of L-proline as the ligand, Cu/CuI as the catalyst with DMSO as the solvent and a reaction time of 40 h. They also reported the coupling of iodobenzene with dibutylamine by using the same conditions and obtained only 10% yield. These results demonstrated that the steric hindrance of acyclic secondary amines inhibited the coupling reaction.

To improve the yields and decrease the reaction times, high-intensity ultrasound was employed by us in the condensation of 2-chlorobenzoic acids with primary and secondary amines using the Ullmann condensation reaction in DMF as the solvent.

There are some examples concerning the use of ultrasound to increase both the reaction rates and the yields of products and allowing, in many cases, milder reaction conditions.[21–26] High-power ultrasound has become the focus of a new field, known as "sonochemistry". It involves the study of the effects of acoustic waves on chemical systems. The cavitation produced by the passage of ultrasonic waves through the solvent gives rise to sufficiently high temperatures and pressures to clean the surface of the metal, which hinders the formation of impurities before and during the reaction.[27]

In our initial screening experiments, we chose the condensation of 2-chlorobenzoic acid with propylamine (Table 1, Entry **3a**), morpholine (Table 1, Entry **8b**) or dipropylamine (Table 1, Entry **15b**) (2.0 equiv.) in DMF with copper powder as the catalyst under high-intensity ultrasound as models to determine suitable reaction conditions.

As illustrated in Figure 3, **3a** was obtained in 84% yield after a reaction time of 25 min. When the condensation was carried out with morpholine, 15 min were necessary to achieve 82% yield of **8b**, whereas in the case of dipropylamine, 20 min were required to obtain 72% yield of **15b**.

Finally, we studied the quantity of K_2CO_3 to be employed in those reactions (Figure 4). It was demonstrated that, for all cases, 1.0 equiv. of base was enough to obtain the best yields. Despite the fact that pH control of the solution is necessary when we use classical heating conditions, in this case the basic pH did not affect the reaction yield under ultrasonic irradiation.

Although the results did not differ significantly at 18, 20, 30 and 35 kHz, more reproducible results were obtained at 20 kHz, and all the experiments were carried out at this frequency. Higher frequencies cause higher rates of molecular motion, which results in greater power losses. If the fre-

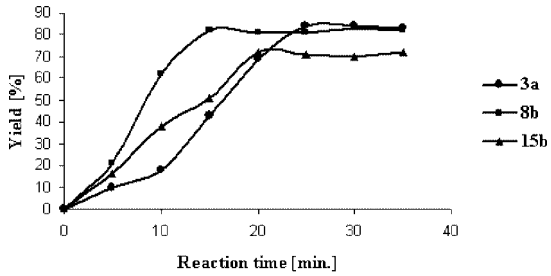


Figure 3. Plot of yield [%] obtained in the condensation of 2-chlorobenzoic acid derivatives with a primary amine (**3a**), secondary cyclic amine (**8b**) or secondary acyclic amine (**15b**) in the presence of ultrasonic irradiation as a function of reaction time [min].

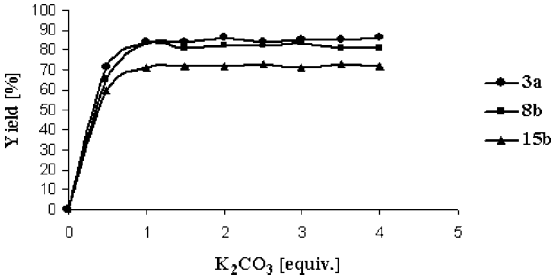


Figure 4. Plot of yield [%] obtained in the condensation of 2-chlorobenzoic acid derivatives with a primary amine (**3a**), secondary cyclic amine (**8b**) or secondary acyclic amine (**15b**) in the presence of ultrasonic irradiation as a function of K_2CO_3 equivalents.

quency is increased, the production of cavities in the liquid is decreased and therefore the sonochemistry effect is less effective.[28–32]

To demonstrate the scope of the ultrasound-assisted method, several 2-chlorobenzoic acids and different primary or secondary amines were used. The results are shown in Table 1. Under these conditions, the yields were significantly improved to 83–97% in primary amines (Table 1, Entries **1a**–**16a**) and to 72–92% in secondary amines (Table 1, Entries **1b**–**20b**). The reaction time was dramatically reduced from hours to minutes.

Conclusions

The use of DMF as the solvent for the condensation of 2-chlorobenzoic acids with aliphatic primary or secondary amines affords the corresponding 2-(alkylamino)benzoic acids or 2-(dialkylamino)benzoic acids in acceptable-to-good yields and with a simple workup procedure. Furthermore, the use of ultrasound-promoted condensation reduces the reaction time in all cases to minutes and improves the yields.

Experimental Section

General: Reactions were monitored by TLC on Merck 60 F_{254} (0.25 mm) plates, which were visualized by CAMAG UV–CABINET II at 254 nm. Melting points were determined in open capillaries with a GALLENKANMP melting point apparatus and are uncorrected. ^1H and ^{13}C NMR spectra were recorded with a Bruker AC 250 F Spectrometer at 300 K and with DMSO as the solvent unless otherwise stated. The ^1H and ^{13}C chemical shifts are reported in ppm relative to residual deuterated solvent signal as an internal reference. The coupling constants (*J*) are given in Hz. Mass spectra were recorded with a Spectrometer Q-TOFII-Micromass, Manchester Ing. Elemental analyses were carried out with a Fisons EA 1108 CHNS-O apparatus at the Microanalytical Unit of the Instituto de Biorgánica de la Universidad de La Laguna, Tenerife, Spain. All reagents purchased from commercial sources were used without further purification. The yields reported in the publication represent an average of at least three independent runs.

Ultrasonic Irradiation Experiments: All ultrasonic irradiation experiments were carried out in a sonochemical apparatus SONIPRED-150. The frequency can be tuned between 18 and 35 kHz and the power can be varied up to a maximum output of 350 W. All the reactions were performed on an open flat-bottomed glass tube (diameter: 30 mm; thickness: 1 mm; volume: 50 mL).

General Procedures for the Condensation of 2-Chlorobenzoic Acids with Primary or Secondary Amines (1a–16a and 1b–20b)

A) Under Conventional Heating: A mixture of the 2-chlorobenzoic acid derivative (0.04 mol), alkylamine (0.08 mol), anhydrous potassium carbonate (0.02 mol) and copper powder (0.003 mol) was heated at reflux in DMF (25 mL) for 2 h (primary amines) or 4 h (secondary amines). The mixture was slowly added with shaking to HCl/H_2O (1:1). The solid obtained was filtered off, washed with water and dissolved in aqueous sodium hydroxide (10%, 30 mL). The basic solution was acidified with $AcOH/H_2O$ (1:3, 40 mL) to pH 5. The 2-(alkylamino)benzoic acid (**1a**–**16a**) or 2-(dialkylamino) benzoic acid (**1b**–**20b**) crystallized, was filtered off and washed with water. Further purification was carried out by recrystallization from $EtOH/H_2O$.

B) Under Ultrasonic Irradiation: The 2-chlorobenzoic acid derivative (0.04 mol), alkylamine (0.08 mol), anhydrous potassium carbonate (0.02 mol) and copper powder (0.003 mol) were added to the flat-bottomed glass tube. The mixture was sonicated for 15–25 min at 20 kHz and 300 W by using an immersion horn. The reactions were monitored by TLC using ethyl acetate/chloroform/acetic acid (8:6:1). After completion of the reaction, the mixture was slowly added with shaking to $HCl:H_2O$ (1:1). The solid obtained was filtered off, washed with water and dissolved in aqueous sodium hydroxide (10%, 30 mL). The basic solution was acidified with $AcOH/H_2O$ (1:3, 40 mL) to pH 5. The 2-(alkylamino)benzoic acid (**1a**–**16a**) or 2-(dialkylamino) benzoic acid (**1b**–**20b**) crystallized, filtered off and washed with water. Further purification was carried out by recrystallization from $EtOH/H_2O$.

The structures of the products obtained were confirmed by melting point, elemental analyses, ^1H NMR and ^{13}CNMR spectroscopy and mass spectra corresponding to those reported in the literature.[16,18,33–36]

Supporting Information (see footnote on the first page of this article): The experimental and spectroscopic data of **1a**–**16a** and **1b**–**20b**.

Acknowledgments

The authors thank Miss Anais Fernández for her kindness in measuring the NMR spectra. We also thank Mr. Juan Carlos Rodriguez for his help with the literature search. Financial support by the Project SECAB-CYTED is gratefully acknowledged.

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Received: March 29, 2007
Published Online: July 3, 2007

