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One-Pot Preparation of Imines from Nitroarenes by a Tandem Process with an Ir–Pd Heterodimetallic Catalyst

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Abstract: A new tandem catalytic process has been studied for a heterodimetallic complex containing both iridium and palladium fragments connected by a 1,2,4-trimethyltriazolyldiylidene ligand. The process implies the unprecedented preparation of imines from the direct reaction of nitroarenes and primary alcohols. The global process comprises the following steps: 1) reduction of the nitroarene to an amine, 2) oxidation of the alcohol to aldehyde, and 3) condensation of the aldehyde and the amine to form the corresponding imine. The oxidation of the alcohol to aldehyde is promoted by the iridium fragment, while the reduction of the nitro group to amine is facilitated by palladium. A wide set of different catalytic systems has been studied, showing that the Ir/Pd complex **1** is a highly active and stable catalyst in the preparation of imines.

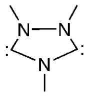
Keywords: alkylation • domino reactions • imines • iridium • nitrogen heterocycles • palladium

Introduction

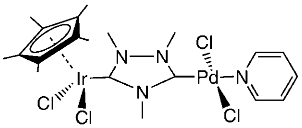
The formation of carbon–nitrogen bonds is one of the most important transformations in organic chemistry.[1] Of particular importance is the N-alkylation of amines by alcohols by the borrowing-hydrogen methodologies.[2,3] Imines are intermediates that are often detected in this synthetic procedures, although their selective preparation has been rarely studied[4,5] Imines are an important class of carbon–nitrogen compounds, because they have a diverse reactivity and multiple applications in laboratory and industrial synthetic processes.[6] The traditional preparation of imines implies the reaction of ketones or aldehydes with amines in the presence of Lewis acid catalysts. They can also be obtained by the oxidative condensation of amines,[7,8] and by oxidation of amines.[8,9] In the past few years, the oxidative coupling of alcohols and amines in the presence of oxygen to provide imines was reported,[5,10,11] but the catalytic outcomes were rather low. Very recently, Milstein and co-workers reported an efficient method for the oxidative coupling of amines and alcohols that produced imines in very high yields.[4]

The selective reduction of nitroarenes is a useful method for the preparation of amines.[12,13] In the study of this reaction, the development of catalytic methodologies that afford high chemo- and regioselectivity under mild reaction conditions is an intriguing area of research.[14]

Over the last few years we have been interested in the formation of carbon-nitrogen bonds through the catalytic coupling of alcohols and amines,,[15,16] via "borrowing-hydrogen" methodologies.[3] We have also been interested in the preparation of stable catalysts that may be compatible with widespread applications,[17] especially using well-defined heterodimetallic complexes to be applied in the design of tandem catalytic reactions.[18,19] In particular, the Ir/Pd complex **1**, in which the two metal centers are linked by the 1,2,4-trimethyltriazolyldiylidene (ditz) ligand, proved to be an efficient catalyst for a series of tandem reactions comprising different transformations of haloacetophenones, typically catalyzed by Ir and Pd.[18] We envisaged that the applications of **1** may be extended to other tandem reactions by combining the large and distinct library of transformations for which Ir and Pd precatalysts are potentially active. In this regard, we now report a general and efficient method for the preparation of imines by the reaction of alcohols and nitroarenes. The reaction implies a tandem process in which a nitroarene is reduced to an aromatic amine by an alcohol,



ditz



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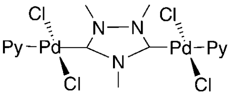
which is oxidized to the aldehyde. The coupling of the aldehyde and the amine selectively affords the corresponding imine. Complexes **2**–**4** were used for comparison purposes.



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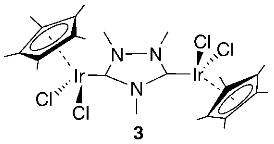

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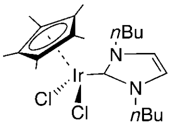

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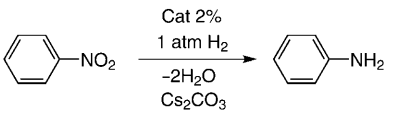
Results and Discussion

In order to determine the ability of a series of homogeneous catalysts toward the reduction of nitroarenes, we first studied the reduction of nitrobenzene with molecular hydrogen. All the reactions were carried out at 100 °C, with 1 atm of H_2. As can be seen from the data shown in Table 1, all the palladium-containing complexes used showed an excellent outcome in this reaction. In contrast, the iridium complexes $[\{\mathrm{IrCp^*Cl_2}\}_2]$ (Cp* = pentamethylcyclopentadienyl) and **3** produced a negligible amount of aniline.

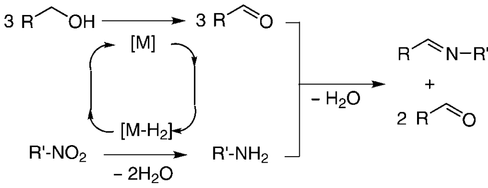
Table 1. Reduction of nitrobenzene to aniline.[a]



Catalyst	t [h]	Aniline [%][b]
$[\{\mathrm{IrCp^*Cl_2}\}_2]$	20	10
$PdCl_2$	10	93
1	10	90
2	10	97
3	20	5

[a] Reaction conditions: nitrobenzene (0.3 mmol), Cs_2CO_3 (0.3 mmol), anisole as internal reference (0.3 mmol), catalyst (2 mol %) in toluene (500 μL). Hydrogen was added with a balloon filled with one atmosphere of gas. Three cycles of vacuum/H_2 were applied before putting the reaction vessel in an oil bath at 100 °C. [b] Yields determined by GC chromatography.

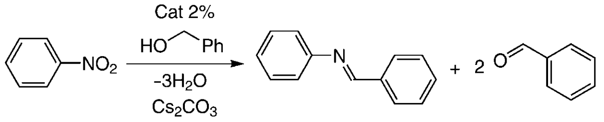
Having proved the ability of the palladium-based complexes to homogeneously catalyze the reduction of nitrobenzene to aniline, and also confirming that the iridium complexes used showed negligible activity in this reaction, we decided to study the direct reaction of nitroarenes and primary alcohols. The process implies the following steps: 1) reduction of a nitroarene to an amine, 2) oxidation of an alcohol to an aldehyde, and 3) condensation of the aldehyde and the amine to form the corresponding imine. This would imply a new tandem reaction comprising two different steps, one of which is catalyzed by palladium (reduction of the nitroarene) and the other by iridium (oxidation of the alcohol to aldehyde).[20,15] The overall reaction, which constitutes a new borrowing-hydrogen process, is disclosed in Scheme 1.



Scheme 1. Tandem reaction studied in this work.

We designed this reaction with the initial idea that an iridium–palladium heterobimetallic complex would be the perfect candidate for the overall process. The iridium fragment would facilitate the alcohol oxidation with the release of hydrogen, while the palladium fragment would enable the nitrobenzene reduction using the hydrogen released during the iridium oxidation process. Table 2 shows the results of the reaction between nitrobenzene and benzylalcohol used as reagent and solvent at 110 °C in the presence of Cs_2CO_3.

Table 2. Catalyst screening for the reaction of nitrobenzene and benzylalcohol.[a]

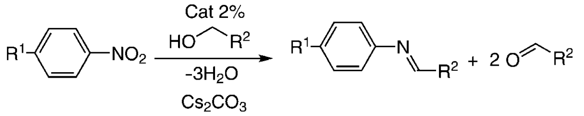


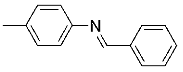
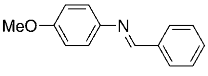
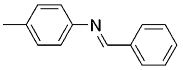
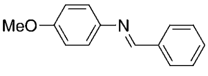
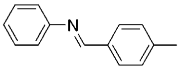
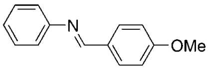
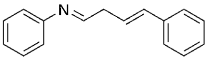
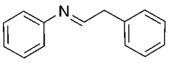
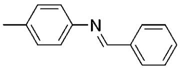
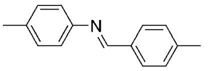
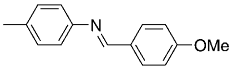
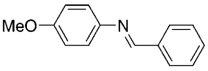
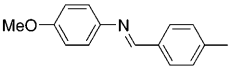
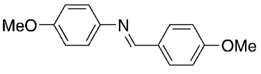
Entry	Catalyst	Cat. [mol %][b]	t [h]	N-Benzylideneaniline [%][c]
1	$[\{\mathrm{IrCp^*Cl_2}\}_2]$	2	3	81
2	$[\{\mathrm{IrCp^*Cl_2}\}_2]$	0.5	20	60
3	$PdCl_2$	2	18	12
4	$[\{\mathrm{IrCp^*Cl_2}\}_2] + PdCl_2$	2	3	72
5	**1**	2	3	87
6	**1**	0.5	20	76
7	**2**	2	3	16
8	**2**	0.5	20	7
9	**3**	2	3	85
10	**3**	0.5	20	35
11	**2**+**3**	0.5	20	73
12	**4**	0.5	20	52

[a] Reaction conditions: nitrobenzene (0.3 mmol), benzylalcohol (5 mmol) used as solvent and reagent, Cs_2CO_3 (0.3 mmol), anisole as internal reference (0.3 mmol). The solution was heated at 110 °C under aerobic conditions. [b] Based on metal amount. [c] Yields determined by GC chromatography.

As can be seen from the results shown in Table 2, all iridium-containing complexes provided good conversions to the imine, while the catalysts that only contain palladium afforded negligible activities (see entries 3, 7, and 8). This result is not surprising, since the palladium catalysts are not expected to oxidize the alcohol to aldehyde and release the required amount of hydrogen to reduce the nitrobenzene to aniline. On the other hand, those catalysts containing only iridium, provide high yields of *N*-benzylideneaniline (entries 1, 2, and 9). This result is remarkable, because it suggests that, for these catalysts, the reduction of the nitrobenzene may be due to a hydrogenation-transfer mechanism, rather than the direct reduction with released molecular hydrogen from the alcohol. The fact that the {IrCp*} fragment may reduce the nitro group by means of a hydrogen transfer process in the presence of *i*PrOH, was previously pointed out by Fujita and co-workers, although the conversions to aniline were very low.[21,22] In a similar experiment, we performed the reduction of nitrobenzene to aniline with *i*PrOH using catalyst **1** (2 mol%). After 20 h, we obtained a quantitative yield of aniline. The Ir–Pd heterodimetallic complex **1** provides the best catalytic outcomes in this reaction (entries 5 and 6), although a mixture of the homodimetallic complexes **2** and **3** (entry 11), also affords good yields. As a consequence of the stoichiometry of the process, in all the reactions under study, benzaldehyde was always detected in an amount over two equivalents with respect to the imine.

Table 3. Catalytic condensation of nitroarenes and primary alcohols.[a]



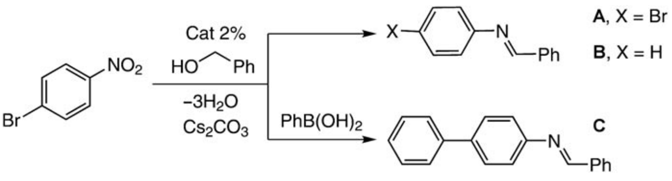
Entry	Catalyst	R^1	R^2	*t* [h]	Product	Yield [%][b]
1	$[\{IrCp^*Cl_2\}_2]$	4-Me	C_6H_5	3		16
2	$[\{IrCp^*Cl_2\}_2]$	4-MeO	C_6H_5	3		32
3	**2+3**	4-Me	C_6H_5	3		83
4	**2+3**	4-MeO	C_6H_5	3		55
5	**1**	H	4-Me-C_6H_5	5		90
6	**1**	H	4-MeO-C_6H_5	20		50 (43)
7	**1**	H	$C_6H_5CH{=}CHCH_2$	20		17
8	**1**	H	$C_6H_5CH_2$	12		92
9	**1**	4-Me	C_6H_5	3		82
10	**1**	4-Me	4-Me-C_6H_5	12		89
11	**1**	4-Me	4-MeO-C_6H_5	12		83
12	**1**	4-MeO	C_6H_5	3		75
13	**1**	4-MeO	4-Me-C_6H_5	12		84
14	**1**	4-MeO	4-MeO-C_6H_5	12		70 (65)

[a] Reaction conditions: nitroarene (0.3 mmol), benzylalcohol (5 mmol) used as solvent and reagent, Cs_2CO_3 (0.3 mmol), anisole as internal reference (0.3 mmol), catalyst (2 mol%). The solution was heated at 110 °C under aerobic conditions. [b] Yields determined by GC chromatography (isolated yields in parenthesis).

To test the general applicability of the process, we decided to study the catalysts previously described in the condensation of different nitroarenes with a series of primary alcohols. The results are shown in Table 3. The reactions were carried out by using a primary alcohol as the reagent and solvent at 110 °C, with a catalyst loading of 2 mol% in the presence of Cs_2CO_3. The catalytic activity of $[\{IrCp^*Cl_2\}_2]$ was very low after 3 h (entries 1 and 2) compared to the other catalytic systems tried in this series of reactions. Longer reaction times afforded better yields to the final imines, although these were never above 60% after 20 h (data not shown in Table 3). As expected, the general activity of the iridium-only-based system is less prone to induce nitro reduction. The mixture of compounds **2** and **3** afforded a very good outcome for the coupling of *p*-nitrotoluene with benzylalcohol (entry 3), but provided only a moderate yield when *p*-nitroanisole was used (entry 4). Catalyst **1** showed a wider applicability, providing very good yields in the reactions of several nitroarenes and benzylalcohols. Some aliphatic primary alcohols were also used (1-hexanol, 1-butanol), but we did not observe the formation of the imines. On the contrast, 2-phenylethanol provided an excellent outcome in the condensation with nitrobenzene (entry 8).

The different catalytic performances of **1** and **3** can be illustrated by the oxidative condensation of *p*-bromonitrobenzene and benzylalcohol. The results are shown in Table 4. Interestingly, each catalyst leads to the formation of a different product. While **3** affords the expected *N*-benzylidene-*p*-bromoaniline (**A**), catalyst **1** provides the dehalogenated imine (**B**). Pd-catalyzed dehalogenations of aryl-halides can take place in *i*PrOH in the presence of a base, such as Cs_2CO_3, and we have also previously observed that this process can be promoted by catalyst **1**. Because the catalytic dehalogenation and the Suzuki–Miyaura reaction are inter-

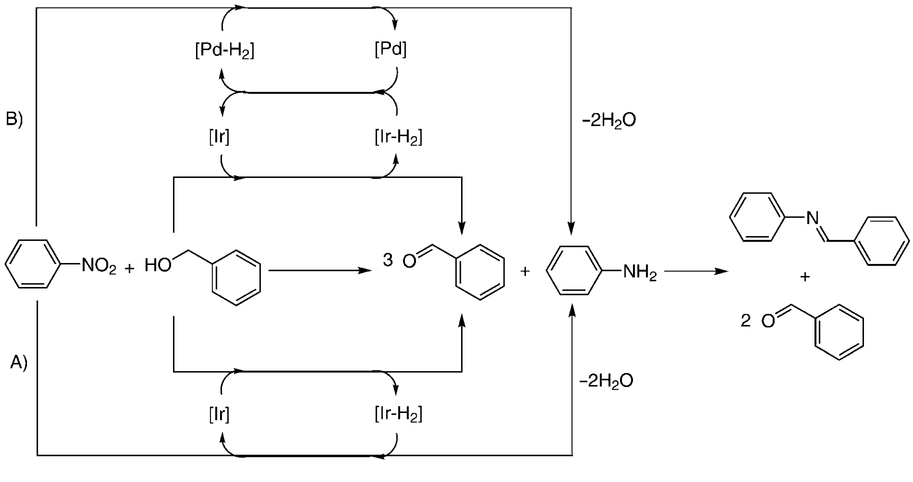
Table 4. Oxidative condensation of *p*-bromonitrobenzene and benzylalcohol.[a]



Entry	Catalyst	Additive	*t* [h]	Product	Yield (%)[b]
1	**3**	none	3	**A**	78 (68)
2	**1**	none	20	**B**	71
3	**1**	$PhB(OH)_2$	40	**C**	77[c] (70)

[a] Reaction conditions: nitrobenzene (0.3 mmol), benzylalcohol (10 mmol) used as solvent and reagent, Cs_2CO_3 (0.3 mmol) and catalyst (2 mol %). $T = 110\,°C$. [b] Yields determined by GC chromatography using anisole as internal standard (isolated yields in parenthesis). [c] Nitrobenzene (0.3 mmol), benzylalcohol (10 mmol), phenylboronic acid (0.3 mmol) Cs_2CO_3 (0.6 mmol) and THF (1 mL).

twined, sharing the oxidative addition step, we decided to combine this reaction with the oxidative condensation of *p*-bromonitrobenzene and benzylalcohol, by adding phenylboronic acid to the reaction mixture. This new reaction provided a very good yield of the corresponding bisarylated imine **C** (Table 4, entry 3), in a reaction in which the two individual actions of the iridium and palladium fragments imply clearly distinct fundamental catalytic processes.



Scheme 2. Two possible mechanistic pathways for the tandem reaction.

Based on these results, we believe that the overall tandem reaction may occur through two different mechanistic pathways (Scheme 2), depending on whether iridium–palladium or “ iridium-only” catalysts are used. For the “ iridium-only” catalysts, the reduction of the nitro group to the amino group only proceeds in good yield for nitrobenzene through the general “borrowing-hydrogen” mechanism (Pathway A, Scheme 2). Based on our experience,[15] the initial stage of the reaction would be the oxidation of the benzyl alcohol to benzaldehyde and the generation of a metal hydride. The metal hydride will reduce the nitrobenzene to aniline. One benzaldehyde equivalent condensates with the generated aniline to produce and hemiaminal intermediate that easily dehydrates to yield the imine. For the Ir/Pd catalysts, an alternative pathway for the reduction of the nitro group could be envisaged. In a first step, the oxidation of the alcohol to aldehyde would be promoted by the iridium fragment generating an iridium hydride. In a second step, an intramolecular hydride migration from iridium to palladium in catalyst **1** would generate a palladium hydride responsible of the nitro to aniline reduction. (Pathway B, Scheme 2).

Conclusion

With this work we have extended the general “borrowing-hydrogen” procedure in the N-alkylation of amines using alcohols. The amine is generated in situ by reduction from a nitroarene. We have evaluated the activity of a series of catalysts in the oxidative coupling of nitroarenes with benzylalcohols to afford the corresponding imines. For this reaction, four homo- and heterodimetallic catalysts of Ir and Pd have been tested. The results provided by the bis(iridium) complexes are remarkable, although limited to several substrates. However, the presence of the palladium fragment in the Ir/Pd complex **1** improves the catalyst activity widening its applicability, most probably because the presence of the Pd opens a new reaction pathway in the reduction of the nitro group by using the hydrogen released in the oxidation of the alcohol by the iridium moiety.

Catalyst **1** has been used for the preparation of different products using the same substrates and changing the additives. In a new illustration of the wide applicability of the Ir/Pd heterodimetallic catalyst, we have also combined the oxidative coupling of *p*-bromonitrobenzene with benzylalcohol, with the Suzuki–Miyaura C−C coupling, by adding phenylboronic acid in the reaction medium. This reaction provides the corresponding bisarylated imine in high yield.

Our work provides a new example of tandem catalysis using dimetallic complexes bearing the 1,2,4-trimethyltriazolyldiylidene ligand. The use of only one catalyst with two different metal fragments is an interesting option for the design of catalytic sequential reactions with fundamentally distinct mechanisms. For these types of processes, a mixture of two catalysts can also be used, but the utilization of a heterodimetallic catalyst provides a series of advantages. We recently suggested that the linkage through the 1,2,4-trimethyltriazolyldiylidene ligand may provide some catalytic cooperativity between the two metals, but other advantages have to be taken into account, as the lower atomic efficiency achieved when two catalysts are used instead of one, and the obvious reduction of waste in the preparation of only one catalyst. Detailed mechanistic studies, development of new heterodimetallic catalysts and new tandem processes are currently in progress in our laboratory.

Experimental Section

General procedures: Compounds **1–4** were prepared according to literature procedures.[18,19,23] All other reagents and solvents were used as received from commercial suppliers. Catalytic experiments were carried out under aerobic conditions and without solvent pre-treatment. Gas chromatograph GC-2010 (Shimadzu) equipped with a FID and a Teknokroma (TRB-5MS, 30 m × 0.25 mm × 0.25 μm) column. Gas chromatograph/Mass spectrometer GCMS-QP2010 (Shimadzu) equipped with a Teknokroma (TRB-5MS, 30 m × 0.25 mm x 0.25 μm) column.

Catalytic studies

Reduction of nitrobenzene to aniline using H_2: Molecular hydrogen was added with a balloon filled with 1 atm of gas to a mixture of nitrobenzene (0.3 mmol), Cs_2CO_3 (0.3 mmol), anisole as internal reference (0.3 mmol) and catalyst (2 mol %) in toluene (500 μL). Three cycles of vacuum/H_2 were applied before putting the reaction vessel in an oil bath at 100 °C for the appropriate time. Yields and conversions were determined by GC chromatography.

Typical catalytic condensation of nitroarenes and primary alcohols: A capped vessel containing a stirrer bar was charged with nitrobenzene (0.3 mmol), benzylalcohol (5.0 mmol), Cs_2CO_3 (0.3 mmol), anisole as internal reference (0.3 mmol), and catalyst (0.5 or 2 mol %). The solution was heated to 110 °C for the appropriate time. Yields and conversions were determined by GC chromatography. Products and intermediates were characterized by GC/MS. Isolated products were characterized by ^{1}H NMR and ^{13}C NMR spectroscopy after column chromatography purification using mixtures of *n*-hexanes/ethylacetate. The confirmation of the nature of the products was performed by comparison with the literature data.[24]

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