

Selenium-Catalyzed Reductive Carbonylation of 2-Nitrophenols to 2-Benzoxazolones

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2-Benzoxazolones or 2-benzimidazolones are synthesized in moderate to good yields in the presence of a base (KOH, NaOH, KOAc, NEt₃, DBU) at atmospheric pressure or under a high pressure of CO by one-pot reductive carbonylation of 2-nitrophenols or 2-nitroaniline in the presence of selenium as catalyst. Besides the effect of base, the effects of solvent and temperature on the reaction were investigated at high

or atmospheric pressure. Contrasting results were obtained for 2-benzoxazolones or 2-benzimidazolone at high and atmospheric pressures. Moreover, phase-transfer catalysis was exhibited.

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Introduction

2-Benzoxazolones and their derivatives are useful intermediates in the production of pharmaceuticals, pesticides, and herbicides.^[1] For example, cyclohexylaminobenzoxazole derivatives are antagonists of *N*-methyl-D-aspartate (NMDA) receptors and are useful for treating stroke, cerebral ischemia, depression, immune diseases etc.^[2] Additionally, 2-benzoxazolone derivatives have found use as achiral templates for enantioselective Diels–Alder reactions,^[3] and chiral oxazolidin-2-ones are widely used as chiral auxiliaries in many important asymmetric syntheses.^[4] These compounds are currently produced by the reaction of phosgene with *o*-aminophenols or *o*-phenylenediamines.^[5] However, because phosgene is highly toxic and corrosive, it is important to develop a non-phosgene reaction.

Several other synthetic methods have also been reported. For example, the carbonylation of *o*-aminophenols or *o*-phenylenediamines with dimethyl carbonate (DMC) affords 2-benzoxazolones or 2-benzimidazolones,^[6] while dialkyl carbonates are obtained from the oxidative carbonylation of alcohols with carbon monoxide.^[7] Clearly, the synthesis of 2-benzoxazolones or 2-benzimidazolones starting directly from carbon monoxide rather than from dialkyl carbonates represents an advantage from the standpoint of atom economy.^[8]

The conversion of 2-aminophenols to 2-benzoxazolones has been achieved by using PdI₂ in conjunction with KI as the catalytic system in 1,2-dimethoxyethane (DME); under the optimized conditions, 2-benzoxazolinone formed with a

yield as high as 96%.^[9] In the carbonylation of α,ω -diamines to 2-imidazolidinones using W(CO)₆ as the catalyst, yields were low.^[10,11] A reaction catalyzed by elemental selenium has been reported to produce high yields, but stoichiometric or excess amounts of selenium are required.^[12–15]

Oxidative carbonylation requires an oxidant, and in most cases the oxidant is oxygen. However, mixing CO and O₂ is dangerous, therefore reductive carbonylation has recently attracted attention owing to the avoidance of using O₂. The transition-metal-catalyzed synthesis of heterocyclic-containing nitrogen compounds by the reductive carbonylation of *o*-substituted nitrobenzenes with carbon monoxide is a well-established method. A heterogeneous, bimetallic Fe–Pd complex catalyzes the carbonylation of 2-nitrophenol under 200 atm pressure of CO with high yields.^[16,17] Additionally, the ruthenium- or palladium-catalyzed reductive carbonylation of *o*-nitrobenzylalcohols gives the corresponding 1,4-dihydro-2*H*-3,1-benzoxazin-2-one derivatives.^[18] 2-Benzoxazolone can also be prepared in high yields by reductive carbonylation of 2-nitrophenol with a high pressure of carbon monoxide and a stoichiometric amount of sulfur or selenium in water in the presence of NEt₃.^[19,20]

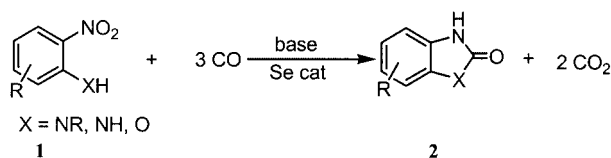
We recently reported a new method for the synthesis of unsymmetrical ureas by direct selenium-^[21] or selenium dioxide-catalyzed^[22,23] reductive carbonylation of nitroaromatics and amines or anilines under 30 atm of CO pressure with the organic base NEt₃ as co-catalyst. In the course of our ongoing investigation of the selenium-catalyzed reductive carbonylation of 2-nitrophenols, we have discovered that the organic bases NEt₃, DBU, and *N*-methylpyrrolidine^[24,25] assist the reaction under high pressures of CO. In this work the carbonylation of 2-nitrophenol or 2-nitroaniline derivatives is studied in the presence of organic (NEt₃, DBU) or inorganic bases (KOH, NaOH, NaOAc, KOAc)

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under atmospheric pressure and low temperature or high temperatures and pressures. A possible reaction pathway of carbonylation was assumed. The results of the detailed investigation are reported here.

Results and Discussion

In a previous paper on selenium-catalyzed reductive carbonylation of 2-nitrophenol,^[24] the products were obtained under high pressure of CO and at high temperature in the presence of organic bases. These were promising but unoptimized results. The system has subsequently been explored more thoroughly. Variables such as the nature of the base, the quantity of base added, CO pressure, solvent, and temperature have now been examined and the results are described below.



The base plays an important role during the reaction. It is well known that organic bases such as NEt_3 and DBU assist the selenium-based catalytic system,^[12–15,21,23–26] but in this work, as shown in Table 1, both NEt_3 and DBU do not demonstrate efficient activity under atmospheric pressure of CO (Table 1, entries 1 and 2), presumably partially due to their high volatility and quick evaporation under atmospheric pressure, resulting in low yields of 2-benzoxazolone (**2a**, X = O). The yields were 3% and 16%, respectively. Inorganic bases such as KOH and KOAc were much more efficient for the reductive carbonylation under atmospheric pressure; the yields were 91% and 93%, respectively (Table 1, entries 3 and 6). For comparison purposes, the experiments were run under identical conditions except for the pressure of CO (Table 1, entries 3 and 16); the yield of 2-benzoxazolone (**2a**) under 30 atm of CO pressure (21%) was much less than that under atmospheric pressure (91%). The choice of solvent also turned out to be significant (Table 1, entries 3, 7, 8 and 10, 13, 14, 15). We observed that selenium is insoluble in toluene when KOH or NEt_3 are used as base under atmospheric pressure of CO. Polar solvents such as DMF and DMSO were essential for the reaction under atmospheric pressure. Under high pressure NEt_3 and DBU with apolar toluene as solvent gave the product in 45% and 57% yields, respectively (Table 1, entries 10, 11, and 12), and the inorganic base KOH was also useful in 38% yield (entry 13), although with DMF as solvent the yields of 2-benzoxazolone were dramatically lower (Table 1, entries 15 and 16).

As seen in Table 2, the yields of **2a** increased as the amount of KOH was increased to six equivalents, and then decreased with any further increase of the amount of KOH. Increasing the amount of added KOH from two equivalents to six equivalents, the yields increased significantly from 25% to 91%. The reaction proceeds very slowly in the presence of a small amount of base (Table 2, entry 1).

Table 1. Effect of base and solvent on the reductive carbonylation of 2-nitrophenol.

Entry	Solvent	Base	Yield ^[a] [%]
1	DMF	NEt_3	<3
2	DMF	DBU	16
3	DMF	KOH	91
4	DMF	NaOH	68
5	DMF	NaOAc	12
6	DMF	KOAc	93
7	DMSO	KOH	86
8	toluene	KOH	–
9	toluene	NEt_3 ^[b]	–
10	toluene	NEt_3 ^[c]	45
11	toluene	DBU ^[d]	57
12	toluene	NEt_3 + DBU ^[e]	67
13	toluene	KOH ^[f]	38
14	DMF	NEt_3	<3
15	DMF	KOH ^[f]	<3
16	DMF	KOH ^[g]	21

^[a] Isolated yields. Reaction conditions: 2-nitrophenol (5.0 mmol), solvent (15 mL), base (6.0 mmol), selenium (0.4 mmol), $T = 110^\circ\text{C}$, $p_{\text{CO}} = 1 \text{ atm}$, $t = 4 \text{ h}$. ^[b] $T = 95^\circ\text{C}$. ^[c] Solvent (10 mL), base (20 mmol), 2-nitrophenol (10 mmol), selenium (0.5 mmol), $T = 150^\circ\text{C}$, $p_{\text{CO}} = 30 \text{ atm}$, $t = 7 \text{ h}$. ^[d] Base (10 mmol); DBU = 1,8-diazabicyclo[5.4.0]undec-7-ene. ^[e] NEt_3 (20 mmol) + DBU (10 mmol). ^[f] Base (6 mmol), $t = 4 \text{ h}$. ^[g] As for [a] except for the pressure of CO. The reaction conditions of entries 11–15 are the same as those of entry 10 except for the note.

Table 2. Effects of the amounts of base, DMF, and the temperature on the reductive carbonylation of 2-nitrophenol under 1 atm of CO pressure.

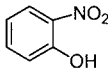
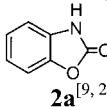
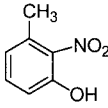
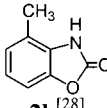
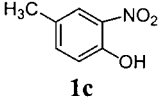
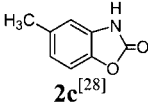
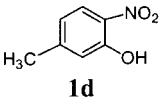
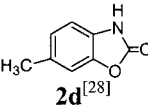
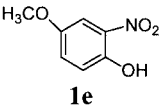
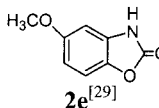
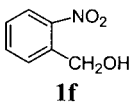
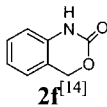
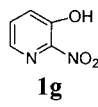
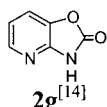
Entry	KOH [mmol]	DMF [mL]	Temp. [$^\circ\text{C}$]	Yield ^[a] [%]
1	2.0	15	110	25
2	3.0	15	110	86
3	6.0	15	110	91
4	10.0	15	110	55
5	3.0	15	125	91
6	3.0	6 ^[b]	95	73
7	3.0	10 ^[c]	95	90
8	3.0	15 ^[b]	95	91
9	3.0	20 ^[b]	95	66
10	3.0	15 ^[b]	80	74

^[a] Isolated yields. Reaction conditions: 2-nitrophenol (5.0 mmol), selenium (0.4 mmol) $t = 4 \text{ h}$. ^[b] $t = 8 \text{ h}$. ^[c] $t = 12 \text{ h}$.

The yield of 2-benzoxazolone was improved by the addition of an excess of DMF (Table 2, entries 6–9). It was necessary to employ dilute solutions under atmospheric pressure of CO to avoid intermolecular reactions. With a fixed amount of catalyst, a more dilute solution did not improve the efficiency of the reaction (Table 2, entry 9). Note that the reactions were run at 80–130 $^\circ\text{C}$ (Table 2, entries 2, 5, 8, and 10). The solvent was evaporated at higher temperature. A low temperature resulted in a slow reaction; for example, the yield of **2a** was 34% at 50 $^\circ\text{C}$ after 8 hours.

Further studies with 2-nitrophenol derivatives **1b**, **1c**, and **1d** probed the effect of the position of methyl substitution on the formation of 2-benzoxazolones (Table 3). In contrast to the results with the substrate **1a**, the yields of **2c** and **2d** were higher under a high pressure of CO, but the yield of **2b** was the same as that of **2a**, which shows that the nearer

Table 3. Reductive carbonylation of 2-nitrophenols under high or atmospheric pressure.

Entry	Substrate	Product	Yield (%)	
			$p^{[a]}_{\text{CO}} = 30 \text{ atm}$	$p^{[b]}_{\text{CO}} = 1 \text{ atm}$
1	 1a	 2a ^[9, 24]	67	91
2	 1b	 2b ^[28]	67	83
3	 1c	 2c ^[28]	78	70
4	 1d	 2d ^[28]	80	61
5	 1e	 2e ^[29]	75	45
6	 1f	 2f ^[14]	87	<5
7	 1g	 2g ^[14]	56	60

^[a] Isolated yields. Reaction condition: toluene (10 mL), 2-nitrophenol (10 mmol), selenium (0.5 mmol), NEt₃ (20 mmol) + DBU (10 mmol), $T = 150^\circ\text{C}$, $t = 7 \text{ h}$. ^[b] 2-Nitrophenol (5 mmol), DMF (15 mL), KOH (6 mmol), selenium (0.4 mmol), $T = 110^\circ\text{C}$, $t = 4 \text{ h}$.

the methyl group is to the nitro group, the lower the yield. This could be because the methyl group causes steric hindrance to the nitro group. In addition, an intermolecular reaction is prone to decrease the yields of 2-benzoxazolones. Under atmospheric pressure of CO, the yields of **2b**, **2c**, and **2d** were lower than that of **2a**. This can be attributed to the electron-donor effect of the methyl group. The results with **1f** show obvious differences between high- and atmospheric-pressure conditions (Table 3). Under atmospheric pressure of CO, **2f** is not easily obtained.^[27] However, selenium catalyzes the reaction of 2-nitrobenzyl alcohols at 30 atm of CO pressure to afford 1,4-dihydro-2H-3,1-benzoxazin-2-ones in good yields.^[25] The reason for this will be investigated in the future.

The results with 2-nitroaniline derivatives under high pressure of CO are shown in Table 4; the yields of 2-benzimidazolones are good to moderate. 2-Benzimidazolone was also obtained under atmospheric pressure with a yield of 57%. Compound **2l** is not easy to obtain under either high- or atmospheric-pressure conditions.

During the reaction process phase transfer of the Se catalyst occurs.^[23,24,26] Before the introduction of CO into the reaction mixture, the catalytic system is heterogeneous and the selenium catalyst is present as a powder. Selenium has a very low solubility in most common solvents, but as the reaction proceeds it dissolves completely and the system becomes a brown homogeneous solution.^[32,33] When the reaction is quenched by stopping bubbling CO and exposing the solution to air, the solution is oxidized to form a grey precipitate of selenium, which can be easily recovered by filtration and can be reused with a yield of 75% to give a catalytic activity similar to that of fresh selenium. That is, the reaction system changes from heterogeneous to homogeneous and then recycles to heterogeneous to allow separation. Therefore, this catalytic system possesses the advantages of both homogeneous and heterogeneous catalysis.

Mechanism

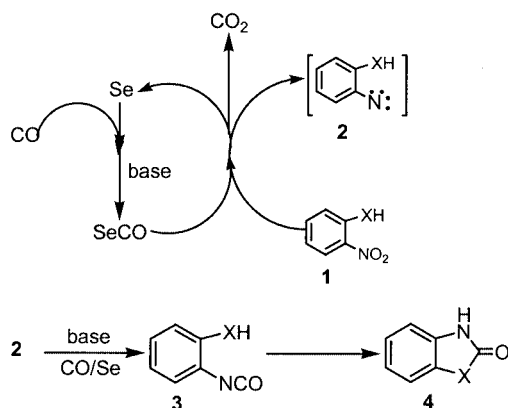
Although a detailed study of the reaction mechanism has not been undertaken, the present reaction can be under-

Table 4. Reductive carbonylation of 2-nitroanilines under high pressure of CO.

Entry	Substrate	Product	Yield (%)
1			92
2			71
3			69
4			66
5			<5

[a] Isolated yields. Reaction conditions: $p_{\text{CO}} = 30$ atm; solvent: toluene; base: NEt_3 ; time: 4 h.

stood mechanistically by assuming the reaction pathway shown in Scheme 1. In the selenium-catalyzed reductive carbonylation of nitroaromatics, it is presumed that the deoxygenation of nitroaromatics **1** with carbonyl selenide (SeCO), generated by the reaction of elemental selenium with carbon monoxide in the presence of base,^[12,13] results in the intermediate nitrene species **2**,^[18,23,34–36] which can react with CO to form isocyanate **3**. This isocyanate reacts further with an amine or hydroxyl group to give the final product **4**.



Scheme 1.

Conclusions

In conclusion, the effect of bases, such as the organic bases NEt_3 , DBU and the inorganic bases KOH and NaOH, on selenium-catalyzed reduction carbonylation of 2-nitrophenol was investigated under atmospheric or high pressure of CO. Contrasting results were obtained for 2-benzoxazolones under high or atmospheric pressure of CO, and the yields were moderate to good. A possible pathway has been postulated. The present one-pot synthesis of 2-benzoxazolones or 2-benzimidazolones represents a new alternative to the use of phosgene.

Experimental Section

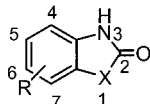
General Remarks: All the organic solvents were of reagent grade and used without further purification. 2-Nitrophenols and 2-nitroanilines, elemental selenium (99.9%), carbon monoxide (99.9%), and base were all used as received. Melting points were determined on a Taike X-4 apparatus (Beijing, China) and are uncorrected. ^1H and ^{13}C NMR spectra were obtained on a Bruker DRX 400-MHz spectrometer. Chemical shifts are reported in ppm relative to tetramethylsilane. $[\text{D}_6]$ Dimethyl sulfoxide was the solvent.

Typical Procedure for the Synthesis of 2-Benzoxazolone. General Procedure for the 2-Nitrophenol Derivatives under High Pressure of CO: Selenium (0.0395 mg, 0.5 mmol), 2-nitrophenol (1.39g, 10 mmol), triethylamine (2.02g, 20 mmol), DBU (1.53g, 10 mmol), and toluene (10 mL) were successively introduced into a 100-mL,

stainless-steel autoclave. The reactor was sealed, flushed with 10 atm of carbon monoxide three times, pressured with 30 atm carbon monoxide, and then placed in an oil bath preheated to 150 °C. After the reaction has finished (7 h), the apparatus was cooled to ambient temperature, and the remaining carbon monoxide was evacuated. The reaction mixture was filtered, and 2-benzoxazolone was collected and further purified by chromatography on silica gel using Et₂O/AcOEt (20:1 to 10:1) as the eluent (yield: 0.90 g, 67%).

Under Atmospheric Pressure: A 50-mL, three-necked flask was fitted with a condenser and charged with 2-nitrophenol (0.70 g, 5 mmol), KOH (0.34 g, 6 mmol), selenium (0.0300 g, 0.38 mmol), and DMF (15 mL). It was then placed in a preheated oil bath at 110 °C and CO (20 mL/min) was bubbled through the reaction mixture with vigorous stirring for 4 h. After cooling to ambient temperature, the resultant mixture was exposed to air and further stirred for about two hours. The mixture was distilled under reduced pressure. The crude product was purified as described above (yield: 0.61 g, 90%).

For 2-nitroanilines, the base was NEt₃ (1.01 g, 10 mmol) and the reaction time was 4 h. Otherwise the procedure was the same as described above for 2-nitrophenol under high pressure of CO.



2-Benzoxazolone (2a): M.p. 137–138 °C (ref.^[9] 137–139 °C). ¹H NMR (400 MHz, [D₆]DMSO, 25 °C): δ = 10.68 (s, 1 H), 7.09–7.31 (m, 4 H) ppm. ¹³C NMR ([D₆]DMSO): δ = 154.50 (C=O), 143.32 (Ph-O), 130.40 (Ph-NH), 123.77 (C-5), 121.84 (C-6), 109.59 (C-7), 109.51 (C-4) ppm.

5-Methyl-3H-benzoxazolone (2c): M.p. 130–131 °C (ref.^[28] 123–125 °C). ¹H NMR (400 MHz, [D₆]DMSO, 25 °C): δ = 11.56 (s, 1 H), 7.15–6.86 (m, 3 H), 2.33 (s, 3 H) ppm. ¹³C NMR ([D₆]DMSO): δ = 154.71 (C=O), 141.41 (Ph-O), 133.41 (Ph-NH), 130.38 (C-5), 122.56 (C-6), 110.11 (C-7), 108.99 (C-4), 20.90 (CH₃) ppm.

1,4-Dihydro-2H-3,1-benzoxazin-2-one (2f): M.p. 118–119 °C (ref.^[14] 118.5–119 °C). ¹H NMR (400 MHz, [D₆]DMSO, 25 °C): δ = 10.19 (s, 1 H), 7.21–6.89 (m, 4 H), 5.28 (s, 2 H) ppm. ¹³C NMR ([D₆]DMSO): δ = 151.95 (C=O), 136.49 (Ph-NH), 128.77 (Ph-CH₂), 124.44, 122.36, 118.58, 113.68, 67.57 (CH₂) ppm.

2-Benzimidazolone (2h): M.p. 298–300 °C (ref.^[14] 300 °C). ¹H NMR (400 MHz, [D₆]DMSO, 25 °C): δ = 10.67 (s, 2 H), 6.96 (m, 4 H) ppm. ¹³C NMR ([D₆]DMSO): δ = 155.43 (C=O), 129.71 (Ph-NH), 120.53 (C-5, C-6), 108.60 (C-4, C-7) ppm. C₇H₆N₂O (134.14): calcd. C 67.68, H 4.43, N 20.88, O 11.93; found C 62.78, H 4.43, N 21.27, O 11.52. IR (KBr): ν̄ = 3025, 1715, 1020, 868, 714 cm⁻¹.

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