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SHORT COMMUNICATION

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Rhodium-Catalyzed Synthesis of 2,5-Epoxybenzo[*f*][1,4]oxazepines by Tandem Reaction of 1-Sulfonyl-1,2,3-triazoles and Salicylaldehydes

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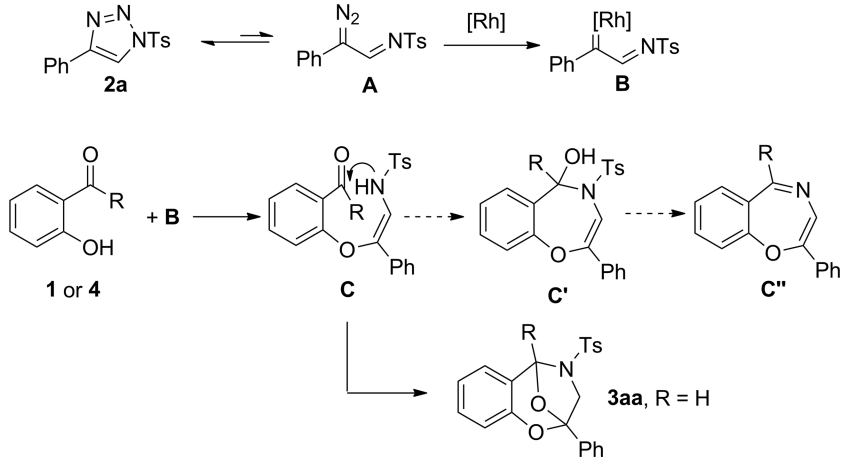
Keywords: Synthetic methods / Homogeneous catalysis / Carbenes / Nitrogen heterocycles / Rhodium / Cyclization

Readily available 1-sulfonyl-1,2,3-triazoles were converted into α-imino carbenes in the presence of catalytic amounts of rhodium(II) salts. The carbenes underwent a tandem reaction with salicylaldehydes to provide a series of functionalized 2,5-epoxybenzo[*f*][1,4]oxazepines in high yields.

Introduction

It is well known that there is a closed/open form equilibrium between 1,2,3-triazole and α-diazo imine species, and the position of this equilibrium mainly depends on the substitution pattern of the triazole.[1] In 2007, the Gevorgyan group used a rhodium(II) catalyst to trap the 2-pyridyl diazo compound, and the corresponding carbenoid underwent cycloaddition with nitrile to furnish a N-fused imidazopyridine.[2] *N*-Sulfonyl-1,2,3-triazole, which is stable and easy to handle, can be readily prepared from terminal alkynes and *N*-sulfonyl azide by copper-catalyzed 1,3-dipolar cycloaddition.[3] In 2008, further improvement by Fokin and Gevorgyan demonstrated that *N*-sulfonyl-1,2,3-triazole can serve as a precursor of α-imino rhodium carbenoid.[4] On the basis of these ideas, many new transformations, such as cycloaddition,[5] cyclopropanation,[6] ring expansion,[7] C–H insertion,[8] heteroatom–H insertion,[9] ylide formation,[10] and so on,[11] have been realized since then. Notably, sometimes the reactivity of α-imino rhodium carbenoids is quite different from that of the corresponding α-oxo carbenes. For example, Murakami realized an intramolecular [3+2] cycloaddition of an aryl ring with α-imino carbenes,[5a] whereas α-diazo esters have been shown to undergo a Büchner reaction to afford bicycle[5.3.0]-deca-1,3,5-trienes. As part of our research interest in carbene-mediated chemical transformations,[12] we realized the synthesis of 3-pyrrolin-2-one,[12a] *N*-acylamidine,[12b] furan derivatives,[12c]



Scheme 1. Initial hypothesis.

and β-(methylthio)-α,β-unsaturated ketones.[12d] On the basis of the achievements of others and our group, we sought to explore cascade reactions involving α-imino rhodium carbenoids. We envisioned that if we were to use salicylaldehyde derivatives to trap the α-imino carbenoid, O–H inser-

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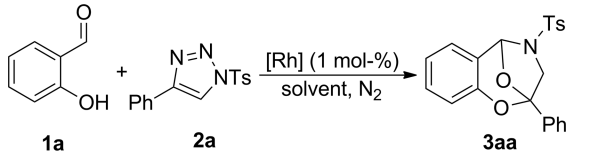
tion would lead to the formation of compound **C** (Scheme 1); subsequent intramolecular nucleophilic addition would produce intermediate **C′**, which may undergo elimination of *p*-toluenesulfonic acid (TsOH)[13] to afford benzo[*f*][1,4]oxazepine **C′′**. To our surprise, upon performing the reaction in the presence of dirhodium(II) tetraoctanoate {[Rh_2(oct)$_4$]}, we obtained the 2,5-epoxybenzo[*f*][1,4]oxazepine **3aa** in high yield. This intriguing result encouraged us to investigate the transformation in detail.

Results and Discussion

The reaction was first conducted in 1,2-dichloroethane (DCE) at 90 °C under a nitrogen atmosphere, and our preliminary investigation focused on the catalyst (Table 1, entries 1–5). Most of the tested rhodium salts gave similar results, whereas [Rh_2{(*S*)-ntv}$_4$] afforded **3aa** in a lower yield (Table 1, entry 5). A slightly higher yield was obtained if toluene was employed as the solvent (Table 1, entry 6), but no products were detected if the reaction was performed in dioxane or MeCN (Table 1, entries 8 and 9). A screen of the reaction temperature revealed that **3aa** could be isolated in 85% yield upon conducting the reaction at 100 °C (Table 1, entry 12).

Table 1. Optimization of reaction conditions.[a]



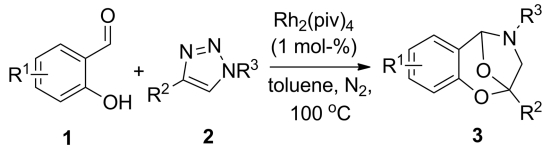
1a **2a** **3aa**

Entry	Catalyst	Solvent	*t* [h]	*T* [°C]	Yield[b] [%]
1	[Rh_2(oct)$_4$]	DCE	2.0	90	73
2	[Rh_2(OAc)$_4$]	DCE	2.0	90	74
3	[Rh_2(piv)$_4$]	DCE	1.5	90	78
4	[Rh_2(esp)$_2$]	DCE	1.5	90	76
5	[Rh_2(*S*-ntv)$_4$]	DCE	3.0	90	66
6	[Rh_2(piv)$_4$]	toluene	1.5	90	80
7	[Rh_2(piv)$_4$]	$CHCl_3$	2.0	70	76
8	[Rh_2(piv)$_4$]	dioxane	11.0	90	–
9	[Rh_2(piv)$_4$]	CH_3CN	11.0	90	–
10	[Rh_2(piv)$_4$]	toluene	9.0	70	55
11	[Rh_2(piv)$_4$]	toluene	3.0	80	78
12	[Rh_2(piv)$_4$]	toluene	1.0	100	85
13	[Rh_2(piv)$_4$]	toluene	1.0	110	75

[a] Triazole **2a** (0.2 mmol) and **1a** (0.24 mmol) were used; the reaction was performed in solvent (2 mL) under an atmosphere of N_2. [Rh_2(piv)$_4$] = dirhodium(II) tetrapivalate, [Rh_2(esp)$_2$] = bis[rhodium(α,α,α′,α′-tetramethyl-1,3-benzenedipropionic acid)]. [b] Yield of isolated products.

We next explored the scope of this transformation under the optimized conditions (Table 1, entry 12). Triazoles bearing sulfonyl groups other than a tosyl group afforded the corresponding products in moderate to high yields (Table 2, entries 1–4). Aryl substrates with an alkyl substituent in the *para* position worked well to give the 2,5-epoxybenzo[*f*][1,4]oxazepines in moderate yields (Table 2, entries 5–7). Substrate **2i**, which contains a more electron-rich aromatic group, was also tested and afforded **3ai** in 70% yield (Table 2, entry 8). Fluoro, chloro, and bromo substituents were all well tolerated, and **3aj**, **3ak**, and **3al** were isolated in yields of 66, 60, and 70%, respectively (Table 2, entries 9–11). Shifting to a trifluoromethyl group led to a much higher yield (Table 2, entry 12). Other functional groups such as cyano and methoxycarbonyl were also compatible with this transformation (Table 2, entries 13 and 14). The use of 4-aryl-1,2,3-triazoles bearing a substituent in the *ortho* or *meta* position of the aryl ring provided the corresponding products in good yields (Table 2, entries 15–19). With respect to the salicylaldehyde derivatives, the reaction also proceeded smoothly to give **3ba**–**ea** in yields ranging from 56 to 80% (Table 2, entries 20–23). Notably, 2-hydroxy-1-naphthaldehyde (**1f**) and 1-(2-hydroxyphenyl)ethanone (**4**) were also successfully incorporated to afford **3fa** and **5** in yields of 72 and 50%, respectively [Equations (1) and (2)].

Table 2. Reaction scope.[a]



1 **2** **3**

Entry	**1** (R^1)	**2** (R^2, R^3)	*t* [h]	Product (yield[b] [%])
1	**1a** (H)	**2b** (Ph, Ms)	1.0	**3ab** (86)
2	**1a** (H)	**2c** (Ph, *p*-$BrC_6H_4SO_2$)	1.0	**3ac** (70)
3	**1a** (H)	**2d** (Ph, *p*-$MeOC_6H_4SO_2$)	1.0	**3ad** (80)
4	**1a** (H)	**2e** (Ph, $TMSCH_2CH_2SO_2$)	1.0	**3ae** (53)
5	**1a** (H)	**2f** (*p*-MeC_6H_4, Ts)	1.0	**3af** (71)
6	**1a** (H)	**2g** (*p*-EtC_6H_4, Ts)	1.0	**3ag** (72)
7	**1a** (H)	**2h** (*p*-*t*BuC_6H_4, Ts)	1.0	**3ah** (77)
8	**1a** (H)	**2i** (*p*-$MeOC_6H_4$, Ts)	1.0	**3ai** (70)
9	**1a** (H)	**2j** (*p*-FC_6H_4, Ts)	1.0	**3aj** (66)
10	**1a** (H)	**2k** (*p*-ClC_6H_4, Ts)	1.0	**3ak** (60)
11	**1a** (H)	**2l** (*p*-BrC_6H_4, Ts)	1.0	**3al** (70)
12	**1a** (H)	**2m** (*p*-$F_3CC_6H_4$, Ts)	1.0	**3am** (83)
13	**1a** (H)	**2n** (*p*-NCC_6H_4, Ts)	1.5	**3an** (62)
14	**1a** (H)	**2o** (*p*-$MeOCOC_6H_4$, Ts)	1.5	**3ao** (52)
15	**1a** (H)	**2p** (*m*-FC_6H_4, Ts)	1.5	**3ap** (60)
16	**1a** (H)	**2q** (*m*-$F_3CC_6H_4$, Ts)	1.0	**3aq** (57)
17	**1a** (H)	**2r** (*m*-$MeOC_6H_4$, Ts)	2.0	**3ar** (73)
18	**1a** (H)	**2s** (*o*-$MeOC_6H_4$, Ts)	2.0	**3as** (50)
19	**1a** (H)	**2t** (2-naphthyl, Ms)	1.5	**3at** (78)
20	**1b** (5-Cl)	**2a** (Ph, Ts)	1.0	**3ba** (64)
21	**1c** (5-Br)	**2a** (Ph, Ts)	1.0	**3ca** (56)
22	**1d** (3-Me)	**2a** (Ph, Ts)	1.0	**3da** (77)
23	**1e** (3,5-*t*Bu)	**2a** (Ph, Ts)	2.0	**3ea** (80)

[a] Triazole **2** (0.2 mmol) and **1** (0.24 mmol) were treated with [Rh_2(piv)$_4$] (0.002 mmol) in toluene (2 mL) under an atmosphere of N_2; Ms = methylsulfonyl. [b] Yield of isolated product.

Compound **3ca** was also synthesized from **3aa** and Br_2, and it underwent smooth Suzuki coupling with $PhB(OH)_2$ to afford compound **6** in 94% yield; the complicated ring system remained intact (Scheme 2). Moreover, upon treat-

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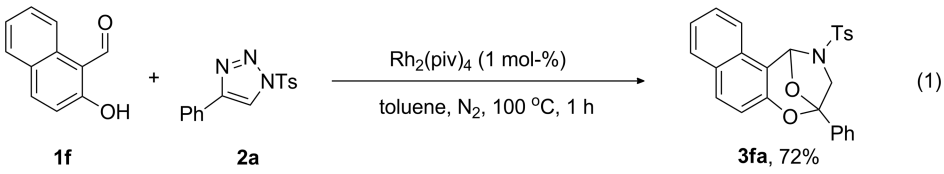
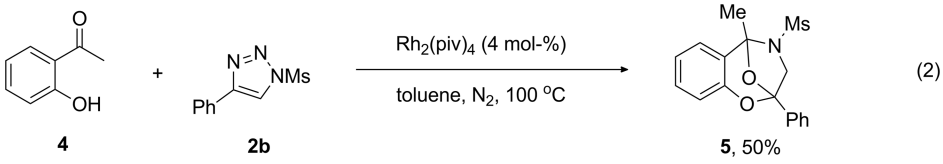
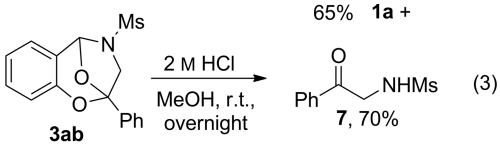
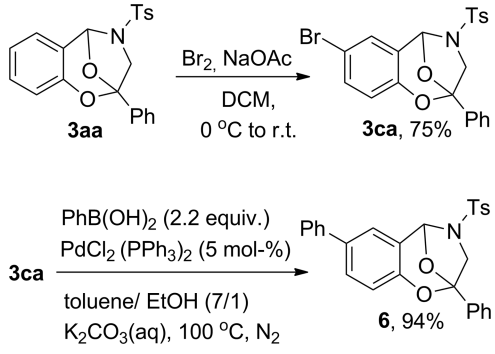




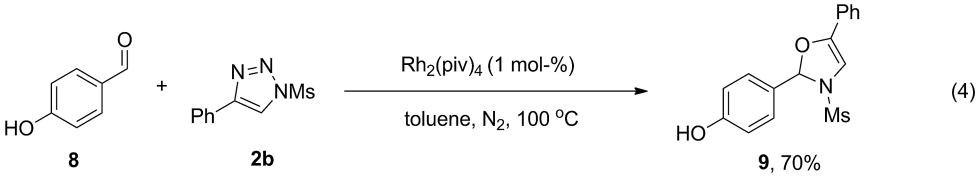


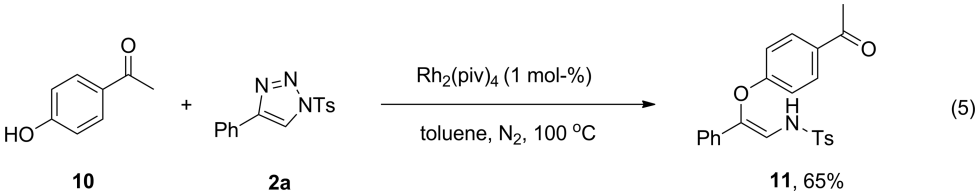


Scheme 2. Synthesis of compound **6**.

ment with 2 M hydrochloric acid, compound **3ab** was hydrolyzed to α-amino ketone **7** in 70% yield [Equation (3)].

Two possible reaction pathways can be envisaged to account for the formation of the 2,5-epoxybenzo[*f*][1,4]oxazepines (Scheme 3). The first mechanistic hypothesis involves O–H insertion, which produces compound **C**. Subsequent stepwise [3+2] cycloaddition and 1,2-shift of the proton lead to the formation of **3** or **5** (Scheme 3, path a). Alternatively, oxazoline **E** can be obtained by formal [3+2] cycloaddition,[14] and it undergoes intramolecular hydroalkoxylation to afford **3** or **5** (Scheme 3, path b). Several control experiments were conducted to shed light on the mechanism. 4-Hydroxybenzaldehyde was used to trap the α-imino rhodium carbenoid, and oxazoline **9** was isolated as the major product in 70% yield [Equation (4)]. However, upon using 4-hydroxyacetophenone (**10**) as the nucleophile, compound **11** was obtained in 65% yield after 1.5 h [Equation (5)]. We also tried to detect intermediate **E** by NMR spectroscopy. In the crude mixture, some characteristic signals could be identified in the ^{1}H NMR spectrum, but isolation of **E** failed, because it hydrolyzed to **1a** and the corre-





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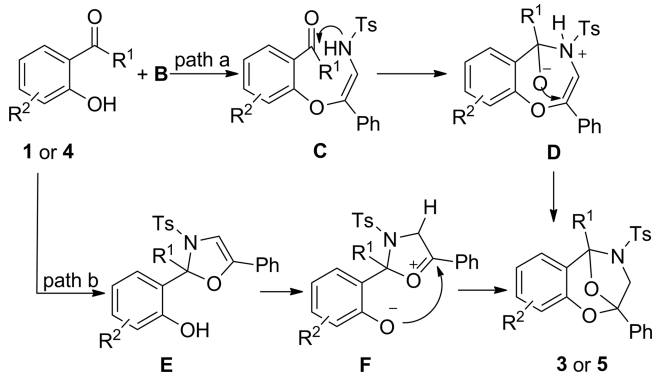
sponding α-amino ketone (see the Supporting Information for more details). On the basis of the above experiments, we believe both pathways are possible and that the mechanism depends on the relative reactivity of the different functional groups.



Scheme 3. Proposed mechanism.

Conclusions

In summary, a novel protocol for the synthesis of 2,5-epoxybenzo[*f*][1,4]oxazepines was developed. The complicated ring system was constructed from readily available starting materials in a one-pot procedure. Many valuable functional groups were well tolerated in this transformation. Considering that 1,4-oxazepine derivatives are important moieties in medicinal chemistry with a variety of biological activities,[15] this protocol may be applicable to the synthesis of related compounds.

Experimental Section

General Procedure: Under a nitrogen atmosphere, toluene (2.0 mL) was added to a reaction flask charged with $[Rh_2(piv)_4]$ (1.2 mg, 1 mol-%), 1-sulfonyl-1,2,3-triazole **2** (0.2 mmol), and *o*-hydroxybenzaldehyde derivative **1** (0.24 mmol) at room temperature. The mixture was then stirred at 100 °C until TLC analysis showed that the unstable intermediate was completely consumed. The mixture was cooled to room temperature and was filtered through a short plug of silica gel. The solution was concentrated and purified by flash chromatography (petroleum ether/EtOAc, 20:1) to give corresponding product **3**.

Acknowledgments

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