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Unsymmetrical Triarylmethanes

1,6-Addition Arylation of *para*-Quinone Methides: An Approach to Unsymmetrical Triarylmethanes

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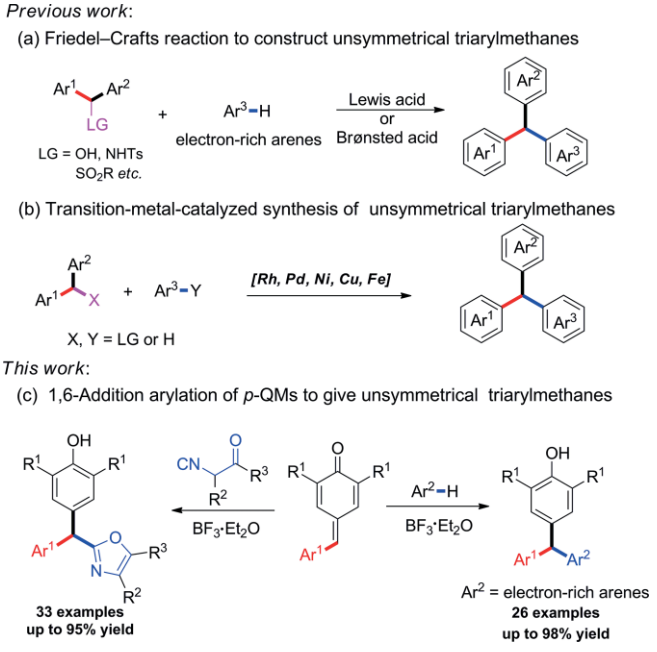
Abstract: A 1,6-addition arylation reaction of *para*-quinone methides with α-isocyanoacetamides and electron-rich aromatic compounds under metal-free conditions has been developed. BF_3·Et_2O plays two roles in the reaction: catalyzing the cyclization of α-isocyanoacetamides to give oxazoles, and activating the *para*-quinone methides to achieve the 1,6-addition arylation process. The reaction shows good functional group tolerance, scalability, and regioselectivity. It is a consice protocol for the synthesis of diverse unsymmetrical triarylmethanes. Further transformation of the resulting triarylmethanes provides an efficient route to some functionalized molecules.

Introduction

The unsymmetrical triarylmethane motif (a triarylmethane with three different aromatic groups) is a privileged structural motif that is widely found in functional materials, pharmaceutical agents, and biologically active molecules.[1] The development of promising strategies for the synthesis of unsymmetrical triarylmethanes has attracted considerable attention in recent decades. Lewis-acid- or Brønsted-acid-catalyzed Friedel–Crafts reactions between nucleophilic electron-rich arenes and unsymmetrical diarylmethanols or related derivatives were the traditional method for the construction of unsymmetrical triarylmethanes (Scheme 1, a).[2] Recently, transition-metal-catalyzed cross-coupling reactions[3] and C–H functionalization[4] have emerged as alternative methods to synthesize these skeletons (Scheme 1, b). Despite this progress, these methods still have shortcomings, including a limited substrate scope, a need for prefunctionalization of the substrates, the use of expensive transition metals, and the formation of undesired regioisomers. This has hampered their wider use to some extent. Thus, the development of a practical method that will improve the reaction efficiency, simplify the operation, and expand the structural scope, using readily available starting materials and an environmentally benign catalyst is still desirable.

Recently, *para*-quinone methides (*p*-QMs), an important and readily available class of synthetic intermediates, have been used to construct diarylmethines through 1,6-conjugate addition.[5,8c] In 2015, Anand's group reported a Pd-catalyzed annulation of *ortho*-alkynylanilines followed by 1,6-addition of *p*-QMs for the synthesis of unsymmetrical triarylmethanes bearing indole groups.[6] Sun's group achieved the synthesis of unsymmetrical triarylmethanes containing pyrrole groups through the chiral-phosphoric-acid-catalyzed 1,6-conjugate addition of in-situ-generated *p*-QMs with 2-methylpyrroles.[7] To construct unsymmetrical triarylmethanes containing more heteroaromatic groups, and also as part of our ongoing interest in *p*-QMs,[8] in this paper we report our results on the BF_3·Et_2O-catalyzed 1,6-addition arylation of *p*-QMs with α-isocyanoacetamides.[9] This results in the direct synthesis of diverse unsymmetrical triarylmethanes with oxazole groups and phenol groups in good to



Scheme 1. Approaches to unsymmetrical triarylmethanes.

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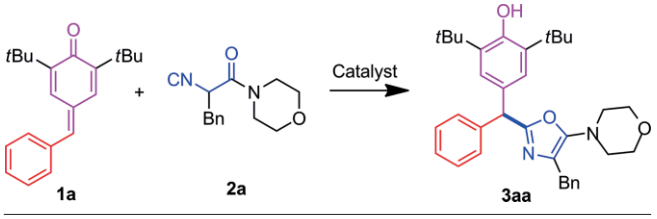
excellent yields under mild conditions (Scheme 1, c). Unsymmetrical triarylmethanes containing both these two groups are important structures that are found in natural products and pharmaceuticals, but the synthesis of this kind of unsymmetrical triarylmethane has never been reported. To further exploit our strategy, we also used dialkyl anilines, methyl indolizine-1-carboxylate, furans, thiophenes, 2-naphthol, and 2-hydroxy-1,4-naphthalenedione as electron-rich aromatic compounds to synthesize other functional unsymmetrical triarylmethanes.

Results and Discussion

Initially, our investigations focused on the combination of *para*-quinone methide **1a** and α-isocyanoacetamide **2a** to optimize the reaction conditions (Table 1). The synthesis of unsymmetrical triarylmethane **3aa** containing a oxazole, a benzene, and a phenol group could be achieved in 60 % yield when BF_3•Et_2O was used as the catalyst (Table 1, entry 9). Other Brønsted or Lewis acids such as AcOH, $(PhO)_2POOH$, $Sc(OTf)_3$, $AlCl_3$, $Cu(OTf)_2$, $Ni(OTf)_2$, $Mg(OTf)_2$, or $Zn(OTf)_2$ gave **3aa** in lower yields (Table 1, entries 1–8). A solvent screening revealed that acetonitrile was better than other solvents, giving **3aa** in 75 % yield (Table 1, entries 10–16). Raising or lowering the reaction temperature did not give a better result (Table 1, entries 17 and 18). By adjusting the equivalent ratio of **1a**/**2a** to 1:2, **3aa** could be finally isolated in 88 % yield (Table 1, entries 19 and 20).

Table 1. Screening of reaction conditions.[a]



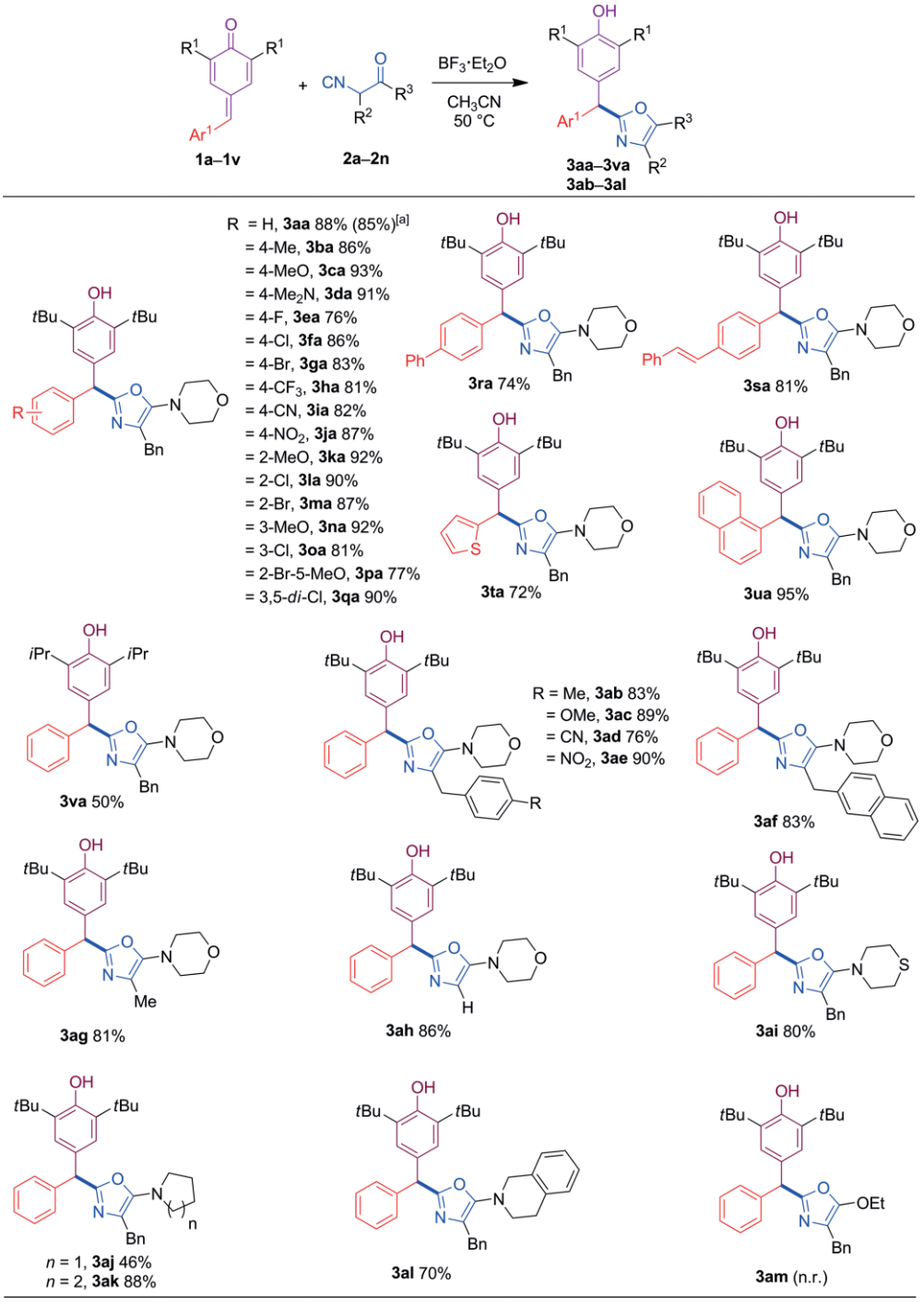
1a 2a 3aa

Entry	Catalyst	Solvent	*T* [°C]	Ratio **1a**/**2a**	Yield [%][b]
1	AcOH	CH_2Cl_2	50	1:1	n.r.
2	$(PhO)_2POOH$	CH_2Cl_2	50	1:1	32
3	$Sc(OTf)_3$	CH_2Cl_2	50	1:1	53
4	$AlCl_3$	CH_2Cl_2	50	1:1	51
5	$Cu(OTf)_2$	CH_2Cl_2	50	1:1	35
6	$Ni(OTf)_2$	CH_2Cl_2	50	1:1	20
7	$Mg(OTf)_2$	CH_2Cl_2	50	1:1	8
8	$Zn(OTf)_2$	CH_2Cl_2	50	1:1	36
9	BF_3•Et_2O	CH_2Cl_2	50	1:1	60
10	BF_3•Et_2O	THF	50	1:1	36
11	BF_3•Et_2O	EtOH	50	1:1	<5
12	BF_3•Et_2O	toluene	50	1:1	14
13	BF_3•Et_2O	$(CH_2Cl)_2$	50	1:1	57
14	BF_3•Et_2O	dioxane	50	1:1	21
15	BF_3•Et_2O	DMF	50	1:1	18
16	BF_3•Et_2O	CH_3CN	50	1:1	75
17	BF_3•Et_2O	CH_3CN	r.t.	1:1	25
18	BF_3•Et_2O	CH_3CN	80	1:1	48
19	BF_3•Et_2O	CH_3CN	50	1:1.5	78
20	BF_3•Et_2O	CH_3CN	50	1:2	93 (88)[c]

[a] Reaction Conditions: **1a** (0.2 mmol), **2a**, and catalyst (0.04 mmol) in solvent (1.0 mL) at the stated temperature for 20 h. [b] The yields were determined by ^{1}H NMR spectroscopy, with dibromomethane as an internal standard. [c] Isolated yields.

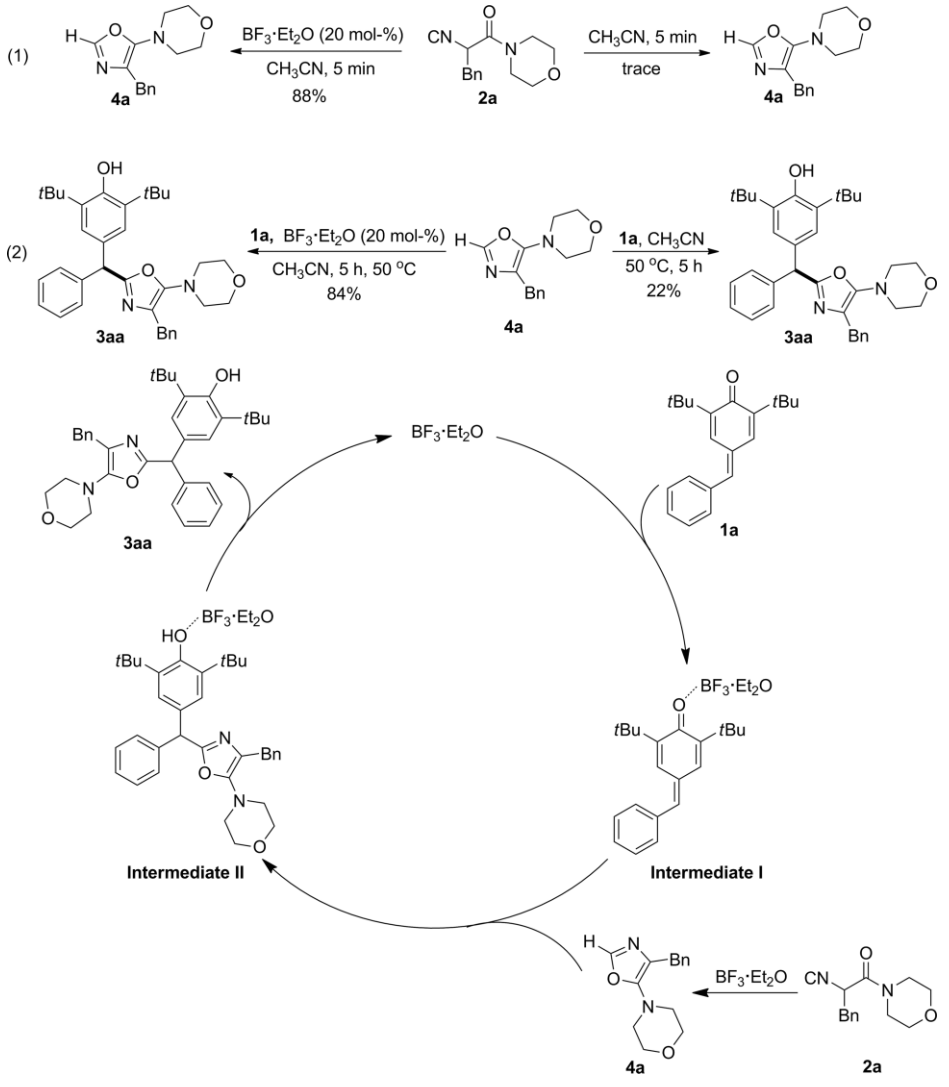
Having established optimized reaction conditions, we explored the substrate scope and generality of this 1,6-addition arylation reaction, and the results are summarized in Scheme 2. *p*-QM derivatives bearing electron-donating groups (-Me, -MeO, $-Me_2N$), halogen atoms (F, Cl, Br), or electron-withdrawing groups ($-CF_3$, -CN, $-NO_2$) at the *para* position of the benzene ring gave **3ba**–**3ja** in 76 to 93 % yields. *p*-QM derivatives with an -OMe, -Cl, or -Br group at the *ortho* or *meta* position gave **3ka**–**3qa** in 77–92 % yields, proving that this reaction is insensitive to the *ortho*, *meta*, and *para* substituents on the phenyl rings. The fact that halogen atoms (Cl, Br) are tolerated in different positions means that further transformation of the unsymmetrical triarylmethane products should be possible. *p*-QM derivatives with phenyl, alkenyl, thiophenyl, and 1-naphthyl groups were also tested, and the corresponding arylated products (i.e., **3ra**–**3ua**) were obtained in 72–95 % yields. Changing the R^1 group of the *p*-QM derivatives to an isopropyl group resulted in the formation of **3va** in 50 % yield. Next, we explored the scope of the reaction with respect to the α-isocyanoacetamide component. Substrates with electron-donating or electron-withdrawing groups at the *para*-position of the benzyl group were well tolerated, giving **3ab**–**3ae** in 76–90 % yields. When R^2 was varied to naphthalen-2-ylmethyl and methyl groups, **3af** and **3ag** were formed in 83 and 81 % yields. Moreover, **3ah**, with the C-4 position of the oxazole ring unsubstituted, was obtained in 86 % yield. When R^3 was changed from a morpholino to a thiomorpholino or another cyclic amino group, **3ai**–**3al** were formed in 46–88 % yield. When α-isocyanoacetamide **2a** was replaced with α-isocyanoacetate, the corresponding product (i.e., **3am**) was not obtained. To probe the efficiency of our reaction, a gram-scale experiment (**1a**: 4 mmol) was carried out. When the catalyst loading was decreased to 15 mol-%, **3aa** was still obtained in 85 % yield (1.84 g).

To explore the detailed mechanism of our reactions, control experiments were carried out as shown in Scheme 3. When the reaction was run without **1a**, α-isocyanoacetamide **2a** was transformed into 4,5-disubstituted oxazole **4a** in 88 % yield in 5 min under standard conditions.[9f] When **2a** was treated without BF_3•Et_2O, only a trace amount of **4a** could be detected [Scheme 3, Equation (1)]. These results indicated that in our system, the cyclization of **2a** to form **4a** occurs before the 1,6-addition of *p*-QMs; this is different from previously reported reactions involving α-isocyanoacetamides, which proceed through nucleophilic addition followed by cyclization. Combining **4a** with **1a** under standard conditions, the arylation pathway proceeded smoothly to give **3aa** in 84 % yield, even when the reaction time was shortened to 5 h. Compound **3aa** was only obtained in 22 % yield in the absence of BF_3•Et_2O [Scheme 3, Equation (2)]. Based on these data, we speculate that BF_3•Et_2O plays two roles in our reaction: it catalyzes the cyclization of α-isocyanoacetamide **2a** to form oxazole **4a**, and it activates *para*-quinone methide **1a** to achieve the 1,6-addition arylation process via intermediates **I** and **II**. A detailed catalytic cycle is proposed in Scheme 3.



Scheme 2. Substrate scope. Reaction conditions: **1** (0.2 mmol), **2** (0.4 mmol), BF_3·Et_2O (0.04 mmol) in acetonitrile (1.0 mL) at 50 °C for 20 h. Isolated yields are given. [a] 4 mmol scale.

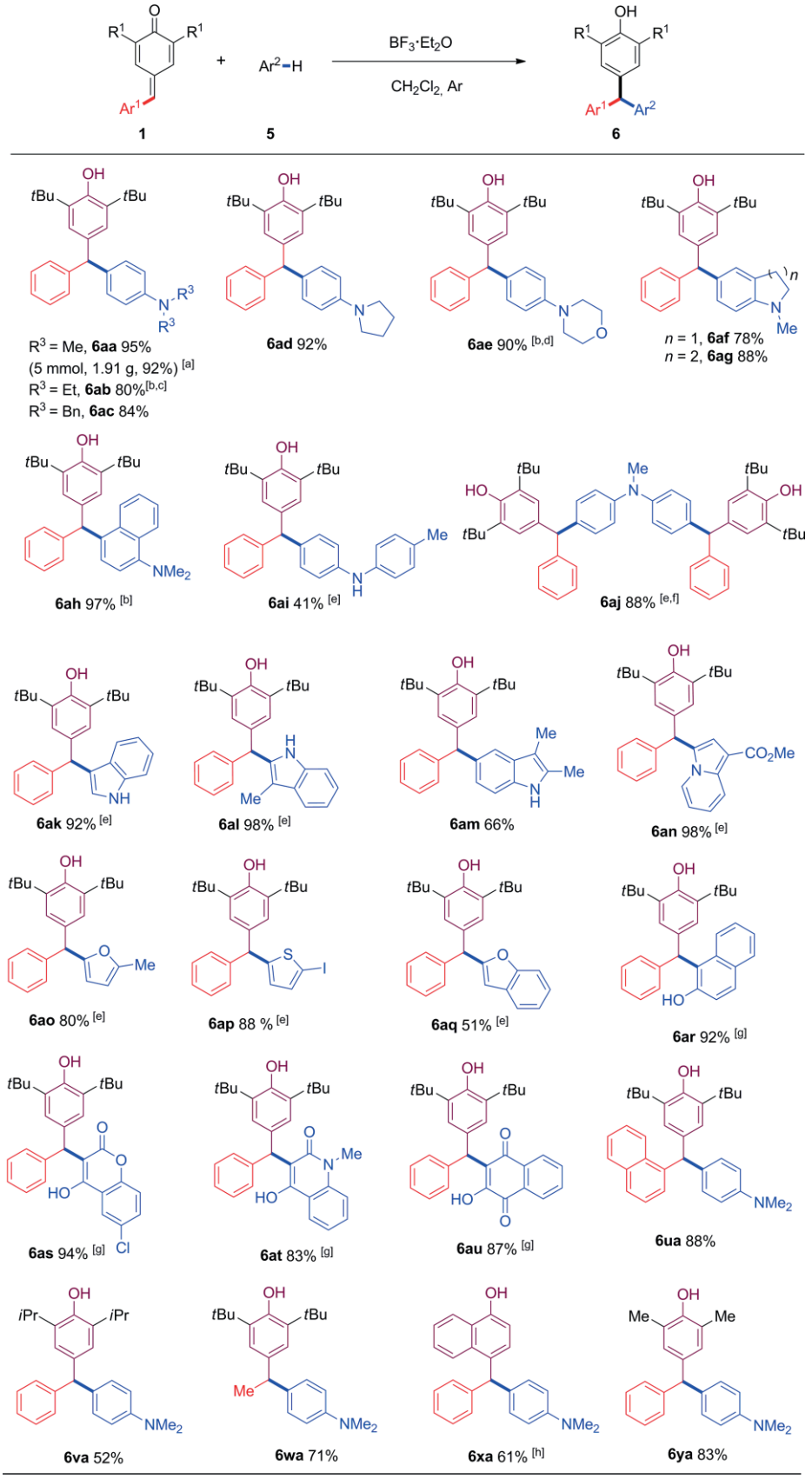
Based on the above reaction mechanism, we supposed that nucleophilic electron-rich aromatic compounds could also be used to construct unsymmetrical triarylmethanes using our catalytic system. Since nitrogen-containing unsymmetrical triarylmethanes are important for dyes[10] and medicinal compounds,[11] various nitrogen-containing aromatic compounds were then tested, and the results are summarized in Scheme 4. Dialkyl anilines **5a**–**5c** gave arylated products **6aa**, **6ab**, and **6ac** in 95, 80, and 84 % yields. Cyclic amines, such as pyrrolidine, morpholine, *N*-methylindoline, and *N*-methyltetrahydroquinoline, gave **6ad**–**6ag** in 78–92 % yields. When *N*,*N*-dimethylnaphthalen-1-amine (**5h**) was examined, **6ah** was obtained in 97 % yield. A substrate bearing a secondary amine gave **6ai** in 41 % yield. When aminodiphenylmethane was tested, double 1,6-addition arylated product **6aj** was assembled quickly in 88 % yield in 5 min; this compound can serve as the core struc-



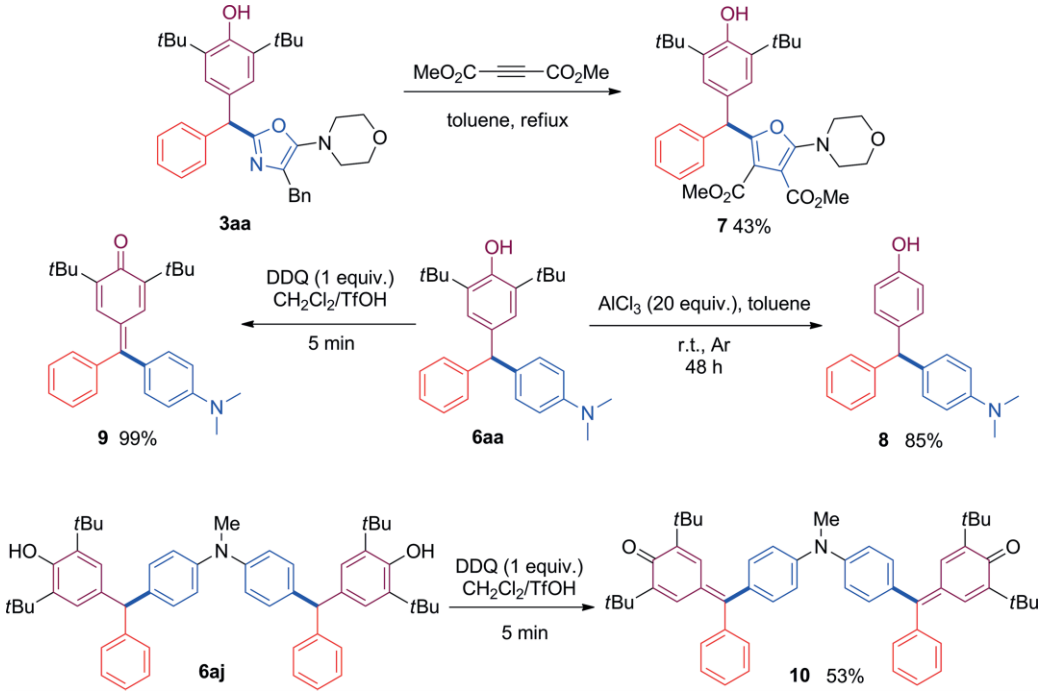
Scheme 3. Control experiments and proposed catalytic cycle.

ture of dendritic architectures. After checking the arylamines, we then turned our attention to other important heteroaromatic compounds. Generally, the reaction proceeded smoothly to give the unsymmetrical triarylmethanes in moderate to good yields at room temperature within short reaction times. Indole and 3-methylindole gave C-3- and C-2-arylated products **6ak** and **6al** in 92 and 98 % yields, while 2,3-dimethylindole gave C-5-arylated product **6am** in 66 % yield. Methyl indolizine-1-carboxylate reacted selectively at the C-3 position to give **6an** in 98 % yield. Other heteroaromatic rings such as furan, thiophene, and benzofuran could also be converted into the desired products (i.e., **6ao**–**6aq**) in good yields. 2-Naphthol, 4-hydroxycoumarin, *N*-methyl-4-hydroxyquinol-2-one, and 2-hydroxy-1,4-naphthalenedione were also used in the reaction, and gave **6ar**–**6au** in good yields from 83 to 92 %. When the phenyl group of the *p*-QM component was changed to a naphthyl or methyl group, the expected products (i.e., **6ua** and **6wa**) were formed in 88 and 71 % yields, respectively. *p*-QM derivatives with isopropyl and methyl groups were also tolerated, and gave target arylated products **6va** in 52 % yield, and **6ya** in 83 % yield. 1-Naphthoquinone 4-methide (*p*-NMQ) also reacted smoothly to give **6xa** in 61 % yield under our reaction conditions. To our delight, when the catalyst loading was lowered to 15 mol-%, compound **6aa** was still formed in 92 % yield on a 5 mmol scale (1.91 g).

With the aim of gaining insight into the utility of the reaction, the arylated product was further transformed. Tetrasubstituted furan **7** was obtained conveniently from **3aa** and an alkyne in 43 % yield through a Diels–Alder reaction (Scheme 5). Furthermore, an $AlCl_3$-promoted de-*tert*-butylation of **6aa** gave compound **8** in 85 % yield at room temperature. In addition, **6aa** and **6aj** could be oxidized with DDQ (2,3-dichloro-5,6-dicyano-1,4-benzoquinone) to give **9** in 99 % yield, and **10** in 53 % yield; these compounds are also core structures of many biologically active molecules,[12] materials for dye-sensitised solar cells (DSC),[13] and chemodosimeters for the selective and chromogenic sensing of cyanide anions.[14]



Scheme 4. Substrate scope. Reaction conditions: **1** (0.2 mmol), **5** (0.24 mmol) and BF_3·Et_2O (0.04 mmol) in CH_2Cl_2 (1.0 mL) at 40 °C for 10 h under Ar. Isolated yields are given. [a] 5 mmol scale with 15 mol-% BF_3·Et_2O as catalyst at 40 °C for 24 h under Ar. [b] 50 °C. [c] 24 h. [d] 120 h. [e] r.t., 5 min. [f] **1a** (0.4 mmol), **5** (0.2 mmol) and BF_3·Et_2O (0.04 mmol) in CH_2Cl_2 (1.0 mL). [g] r.t., 24 h. [h] 40 °C, 24 h.



Scheme 5. Transformation of triarylmethanes.

Conclusions

In conclusion, we have developed a BF_3·Et_2O-catalyzed 1,6-addition arylation of *para*-quinone methides (*p*-QMs) with α-isocyanoacetamides. The reaction allows the direct synthesis of unsymmetrical triarylmethanes containing both oxazoles and phenols in good to excellent yields under mild conditions for the first time. Moreover, our method was extended to other electron-rich aromatic compounds. This metal-free arylation strategy showed good functional-group tolerance and scalability up to gram scale. Transformation of the unsymmetrical triarylmethane products further increased the range of accessible compounds, and revealed potential applications of the products.

Experimental Section

1,6-Addition Arylation of *para*-Quinone Methides (*p*-QMs) with α-Isocyanoacetamides: *p*-QM **1** (0.2 mmol, 1.0 equiv.) and substituted α-isocyanoacetamide **2** (0.4 mmol, 2.0 equiv.) were dissolved in CH_3CN (1.0 mL), and then BF_3·Et_2O (5.1 μL) was added. The reaction mixture was vigorously stirred under air at 50 °C. After the reaction was complete (detected by TLC), the solvent was removed under reduced pressure. The crude mixture was purified by flash column chromatography on silica gel to give pure product **3**.

1,6-Addition Arylation of *para*-Quinone Methides (*p*-QMs) with Electron-Rich Aromatic Compounds: *p*-QM **1** (0.2 mmol, 1.0 equiv.) and substituted arylamine or heteroaromatic compound **5** (0.24 mmol, 1.2 equiv.) were dissolved in CH_2Cl_2 (1.0 mL), and then BF_3·Et_2O (5.1 μL) was added. The reaction mixture was vigorously stirred under argon at room temperature to 50 °C for the appropriate time. After the reaction was complete (detected by TLC), the solvent was removed under reduced pressure. The crude mixture was purified by flash column chromatography on neutral alumina to give pure product **6**.

Acknowledgments

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Keywords: Synthetic methods · Polycycles · Carbocycles · Triarylmethanes

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