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Spiroheterocycles

Synthesis of Dinitrogen-Fused Spirocyclic Heterocycles via Organocatalytic 1,3-dipolar Cycloaddition of 2-Arylidene-1,3-indandiones and an Azomethine Imine

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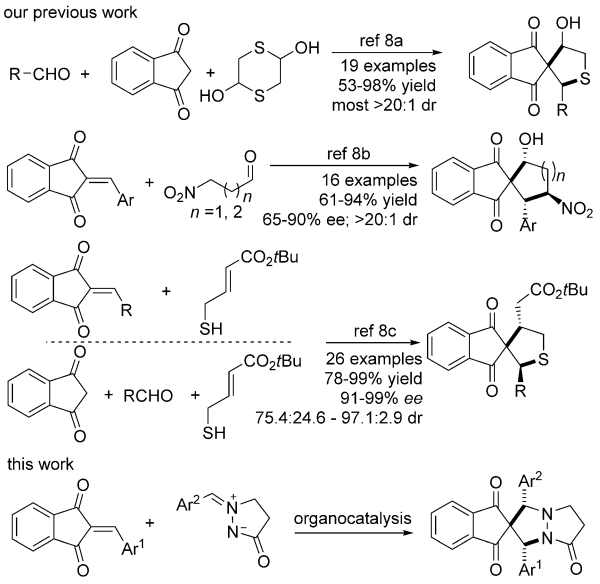
Abstract: An efficient 1,3-dipolar cycloaddition of 2-arylidene-1,3-indandiones with an azomethine imine has been developed to furnish spiroindane-1,3-dione-pyrazolidinones in generally good to high yields with excellent diastereoselectivity under mild conditions.

Introduction

Heterocycles are privileged structural units that are frequently encountered in biologically active natural products as well as in pharmaceuticals and agrochemicals.[1] In particular, dinitrogen-fused heterocycles are the core moieties in many biologically active compounds and have been widely investigated as herbicides, pesticides, and analogues of β-lactam antibiotics, such as penicillin and cephalosporin.[2] Therefore, practical and efficient methods for the synthesis of dinitrogen-fused heterocycles have attracted much attention.[3] Among various methods, metal-catalyzed and organocatalytic 1,3-dipolar cycloadditions based on azomethine imines stand out, and have been extensively investigated.[4] Specially, using *N,N*-cyclic azomethine imine as synthon provides efficient access to dinitrogen-fused heterocycles.[5,6] Notably, several catalyst-free 1,3-dipolar cycloadditions of *N,N*-cyclic azomethine imines proceeded smoothly to furnish dinitrogen-fused heterocycles.[7]

Recently, readily accessible 2-arylidene-1,3-indandiones have been widely used for the synthesis of spiroindane-1,3-dione skeletons,[8] which are important scaffolds in medicinal chemistry[9] and also serve as versatile building blocks for the construction of many bioactive natural products, drugs, and therapeutic leads.[10] Due to the biomedical importance of dinitrogen-fused heterocycles and spiroindane-1,3-dione skeletons, the combination of these two units in one molecule benefits diversity in structure and drug discovery. However, to our knowledge, 1,3-dipolar cycloaddition reactions of 2-arylidene-1,3-indandiones with 1,3-dipoles have not been reported. We aimed to developing an efficient, organocatalytic 1,3-dipolar cycloaddition between 2-arylidene-1,3-indandiones and azomethine imines for the construction of complex polycycles containing two bioactive units; a dinitrogen-fused heterocycle and a spiroindane-1,3-dione.

Based on our previous work and as a continuation of our research interests in synthesis of multicyclic spiro-1,3-indandiones,[8] here we disclose an organocatalytic 1,3-dipolar cycloaddition of 2-arylidene-1,3-indandiones with *N,N*-cyclic azomethine imines to afford highly functionalized spiroindane-1,3-dione-pyrazolidinones in good yields with excellent diastereoselectivity under mild conditions (Scheme 1).



Scheme 1. Synthesis of spiroindane-1,3-dione skeletons.

Results and Discussion

We initiated our investigation by evaluating the reaction of 2-benzylidene-1,3-indandione **1a** and azomethine imine **2a** in the presence of 5 mol% of Et_3N with CH_2Cl_2 as the reaction medium. To our gratification, the cycloaddition proceeded efficiently to provide the spiroindane-1,3-dione-pyrazolidinone **3aa** in 89% yield with excellent diastereoselectivity (>20:1dr)

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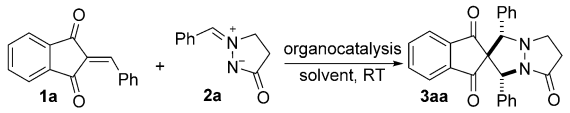
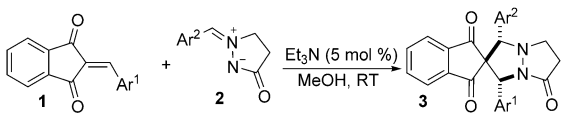
Table 1. Optimization of reaction conditions.



Entry[a]	Catalyst	Solvent	*t* [h]	Yield [%][b]	dr[c]
1	Et_3N	CH_2Cl_2	48	89	>20:1
2	DBU	CH_2Cl_2	48	70	>20:1
3	DABCO	CH_2Cl_2	72	74	>20:1
4	K_2CO_3	CH_2Cl_2	96	71	>20:1
5	Et_3N	THF	72	72	>20:1
6	Et_3N	MeCN	72	75	>20:1
7	Et_3N	DMSO	48	trace	>20:1
8	Et_3N	EtOAc	96	86	>20:1
9	Et_3N	MeOH	48	98	>20:1
10[d]	Et_3N	MeOH	48	94	>20:1
11[e]	–	MeOH	48	21	>20:1

[a] Unless noted, **1a** (0.3 mmol), catalyst (5 mol%), and **2a** (0.36 mmol) in solvent (2 mL) was stirred at room temperature for the time given. [b] Yield of isolated product. [c] Determined by ^{1}H NMR spectroscopy of the crude mixture. [d] Et_3N (1 mol%) was used. [e] Et_3N (0 mol%) was used. DABCO = 1,4-diazabicyclo[2.2.2]octane; DBU = 1,8-diazabicycloundec-7-ene.

Table 2. Substrate scope of 2-arylidene-1,3-indandiones.



Entry[a]	Ar^1	Ar^2	*t* [h]	**3**, yield [%][b]	dr[c]
1	Ph	Ph	48	**3aa**, 98	>20:1
2	4-FPh	Ph	48	**3ba**, 75	>20:1
3	4-ClPh	Ph	48	**3ca**, 85	>20:1
4	4-BrPh	Ph	48	**3da**, 84	>20:1
5	4-CF_3Ph	Ph	48	**3ea**, 92	4:1
6	4-MePh	Ph	48	**3fa**, 83	>20:1
7	3-BrPh	Ph	48	**3ga**, 65	>20:1
8	3-NO_2Ph	Ph	72	**3ha**, 71	>20:1
9	3-MePh	Ph	48	**3ia**, 93	>20:1
10	3-MeOPh	Ph	48	**3ja**, 82	7:1
11	2-thienyl	Ph	72	**3ka**, 70	>20:1
12	Ph	4-BrPh	72	–	–
13	Ph	4-MePh	72	–	–
14	*i*Pr	Ph	72	–	–

[a] Unless noted, **1** (0.3 mmol), Et_3N (5 mol%) and **2** (0.36 mmol) in MeOH (2 mL) was stirred at room temperature for the time given. [b] Yield of isolated product. [c] Determined by ^{1}H NMR spectroscopy of the crude mixture.

within 48 h (Table 1, entry 1). Encouraged by this promising result, several other bases were screened and Et_3N was found to be most suitable for the annulation (Table 1, entries 2–4). Subsequent investigations on the reaction medium showed that 1,3-dipolar cycloaddition is highly dependent on the solvent used (Table 1, entries 5–9). It was found that the polar protic solvent MeOH was the most suitable solvent for this transformation in terms of both the reaction rate and the yield, affording the desired product in 98% yield with excellent diastereoselectivity (>20:1dr) in 48 h. Particularly, reducing the catalyst loading from 5 mol% to 1 mol%, spiroheterocycle **3aa** was also obtained in 94% yield (Table, entry 10). Without catalyst, the 1,3-dipolar cycloaddition proceeded sluggishly to furnish **3aa** in 21% yield after 48 h but the diastereoselectivity remained at a high level (Table, entry 11).

Having established the optimum reaction conditions, a variety of 2-arylidene-1,3-indandiones **1** with azomethine imines **2** were tested to explore the reaction scope. As shown in Table 2, the 1,3-dipolar cycloaddition is considerably general and tolerates 2-arylidene-1,3-indandiones bearing either electron-rich or electron-deficient groups at the aromatic ring. Moreover, there was no significant electronic effect from the substituents on the aromatic moiety of **1**. It should be noted that all the reactions proceeded smoothly to furnish the expected products **3** in good to high yields of 65–98% with excellent diastereoselectivity for most products of >20:1 dr. 2-(4-(trifluoromethyl)benzylidene)-2*H*-indene-1,3-dione (**1e**) reacted with **2a** to yield the product **3ea** with a diastereomeric ratio of 4:1 (Table 2, entry 5), and product **3ja** with a diastereomeric ratio of 7:1 was obtained from the cycloaddition between 2-(3-methoxybenzylidene)-2*H*-indene-1,3-dione (**1j**) and **2a** (Table 2, entry 10). It should be noted that thiophene-substituted 1,3-indandione **1k** readily participated in this reaction, giving rise to the desired product **3ka** in 70% yield with more than 20:1 dr (Table 2, entry 11).

The scope was, however, restricted to azomethine imine **2a**. It was found that 1,3-dipolar cycloaddition between 2-benzylidene-1,3-indandione **1a** and azomethine imines with aromatic substituents **2** resulted in an intractable mixture of products no matter whether Et_3N was used as catalyst or the reaction was conducted under catalyst-free conditions (Table 2, entries 12 and 13). 2-Alkylidene-1,3-indandione was also incompatible with reaction system (Table 2, entry 14).

The structure of cyclization product **3fa** was unambiguously confirmed by single-crystal X-ray analysis (Figure 1).[11] Two aromatic groups were found to be in the same side of the newly formed five-membered ring. Based on the relative configuration of **3fa**, possible models for the intermediates in the 1,3-dipolar cycloaddition reaction were proposed. As depicted in Scheme 2, the cycloaddition was thought to be a concerted reaction. 2-(4-methylbenylidene)-1*H*-indene-1,3(2*H*)-dione (**1f**) interacts with azomethine imine **2a** via π–π stacking. The ni-

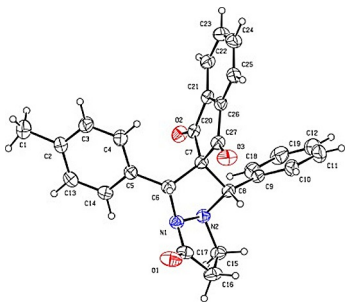
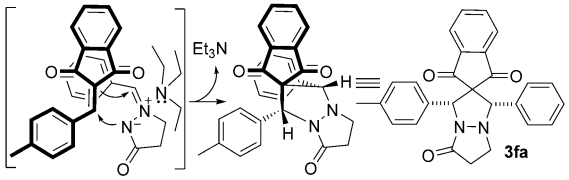


Figure 1. X-ray structure of product **3fa**.



Scheme 2. Possible reaction models in [3+2] cycloaddition.

tranion attacks the less hindered, less substituted end of the double bond while the more substituted end of the double bond reacts with carbon atom of the imine ion to give the desired cycloaddition product **3 fa**. Introducing a substituent into the benzene ring of azomethine imine **2** weakened the interaction between 2-arylidene-1,3-indandiones **1** and azomethine imine **2** as a result of increasing steric hindrance. This might be the reason that azomethine imines with substituted aromatic rings were not compatible with the transformation. Nevertheless, the exact catalytic mechanism of the cycloaddition still needs more investigation.

Conclusions

In summary, we have developed a Et_3N-catalyzed 1,3-dipolar cycloaddition of 2-arylidene-1,3-indandiones with an *N,N*-cyclic azomethine imine to furnish spiroindane-1,3-dione-pyrazolidinones in generally good to high yields with excellent diastereoselectivity under mild conditions. This method tolerates a wide range of 2-arylidene-1,3-indandiones and is a reliable method for the rapid construction of valuable dinitrogen-fused heterocycles. Further investigations of the development of a catalytic asymmetric variant of this reaction and the application of this catalytic approach are currently underway in our laboratory.

Experimental Section

General procedure for the synthesis of spiroindane-1,3-dione-pyrazolidinones 3: To a stirred mixture of 2-arylidene-1,3-indandiones **1** (0.3 mmol) and azomethine imine **2** (0.36 mmol) in MeOH (2 mL) was added Et_3N (5 mol%) and the mixture was kept at room temperature for the time given and monitored by TLC. After removal of the solvent, the crude residue was purified by column chromatography (petroleum ether/ethyl acetate = 3:1, v/v) on silica gel to give the corresponding products **3**.

NMR data of a representative compound: 1′,3′-Diphenyl-6′,7′-dihydro-1′*H*-spiro[indene-2,2′-pyrazolo[1,2-*a*]pyrazole]-1,3,5′(3′*H*)-trione (**3 aa**): ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.97 (d, J = 7.6 Hz, 1H), 7.72–7.68 (m, 1H), 7.58–7.55 (m, 1H), 7.38 (d, J = 7.6 Hz, 1H), 7.19–7.18 (m, 2H), 7.16–7.06 (m, 8H), 5.81 (s, 1H), 4.42 (s, 1H), 3.85–3.79 (m, 1H), 3.10–3.02 (m, 2H), 2.94–2.87 ppm (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 198.3, 194.6, 172.5, 143.1, 141.3, 136.4, 135.6, 135.2, 131.6, 128.8, 128.5, 128.4, 127.8, 127.7, 125.7, 123.3, 123.2, 77.4, 72.1, 63.3, 48.2, 32.5 ppm.

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Keywords: 2-arylidene-1,3-indandiones · azomethine imines · 1,3-dipolar cycloaddition · organocatalysis · triethylamine

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[11] CCDC 1439843 (**3 fa**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre

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