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Catalytic Annulation of Diethyl Methylenecyclopropane-1,1-dicarboxylate with 1,1-Dicyanoalkenes

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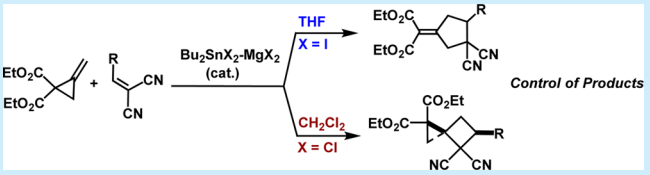
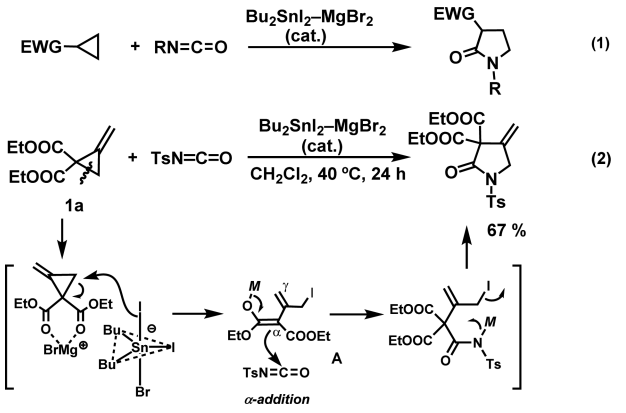
Supporting Information

ABSTRACT: The catalytic annulation of methylenecyclopropane **1** with 1,1-dicyanoalkenes **2** using a Mg−Sn catalytic system was developed. Selective formation of cyclopentylidenemalonates **3** and spiro[2,3]hexane-1,1-dicarboxylates **4** was accomplished via the choice of a proper solvent and an effective catalytic system.



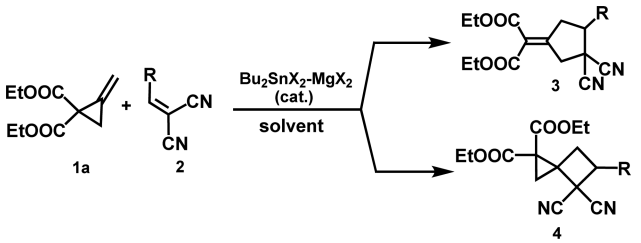
Methylenecyclopropanes (MCPs) can undergo a variety of reactions facilitated by the release of the intramolecular strain of the small ring and its exocyclic C=C bond.[1] The main-group metal and Lewis acid (LA) catalysts activate MCPs, leading to cleavage of the distal bond.[2] Thus, MCPs react not only at the α-position but also at the γ-position to give a variety of cyclic products.[2d] We have established the [3 + 2]-annulation of simple cyclopropanes with isocyanates (Scheme 1, eq 1).[3] We previously reported the formation of an ate

Scheme 1. Catalytic Annulation of Cyclopropanes with Isocyanates



complex, $MgBr^+[Bu_2SnBrI_2]^-$, that serves as the active catalyst in the Bu_2SnI_2−$MgBr_2$ catalytic system. A characteristic of the tin ate complex is its hybrid makeup, wherein both an acidic magnesium cation and a nucleophilic Sn−I bond are parts of the same molecule.[4] Although a similar catalytic principle has been reported for MgI_2,[5] the hybrid characteristic of the tin catalyst confers a higher level of activity. The accessibility of a variety of cyclopropanes is another positive factor for the tin catalyst. We have already applied the tin catalytic system to the reaction of methylenecyclopropane **1a** with an isocyanate, and 4-methylene-γ-butyrolactam was obtained as a product (Scheme 1, eq 2).[3] The ring opening of **1a** afforded tin dienolate **A**, which then reacted with the isocyanate. Intramolecular alkylation of the resultant adduct finally produced a methylene γ-lactam, thus indicating that the initial addition of the isocyanate occurred at the α-position of dienolate **A**. On the basis of these results, herein we present the catalytic reaction of MCP **1a** with dicyanoalkenes **2**, in which magnesium and tin systems combine as effective catalysts. In particular, regioselectivity control of cyclopentylidenemalonates **3** and spiro-[2,3]hexane-1,1-dicarboxylates **4** was accomplished via the proper choice of catalyst and solvent (Scheme 2).

Scheme 2. Catalytic Annulation of Methylenecyclopropanes with 1,1-Dicyanoalkenes



Initially, we performed the catalytic annulation of methylenecyclopropane-1,1-dicarboxylate ester **1a** with 1,1-dicyanoalkene **2a** at 40 °C using THF as solvent (Table 1). In the absence of catalyst, the reaction did not proceed (Table 1, entry 1). There was no reaction when 0.1 equiv of Bu_2SnI_2 was used as the catalyst (Table 1, entry 2). On the other hand, the use of $MgBr_2$ was shown to promote the reaction of methylenecycloprane **1a** and dicyanoalkene **2** (Table 1, entry 3), affording a mixture of cyclopentylidenemalonate **3a** and adduct **4a** in 81% overall yield. MgI_2 as the catalyst afforded **3a** predominantly (Table 1, entry 4). Using a system that combined Bu_2SnI_2 with MgI_2 gave **3a** with the highest level of selectivity (80:2; Table 1, entry 5). In the initial stage, the byproduct **4a** seemed to be a [2

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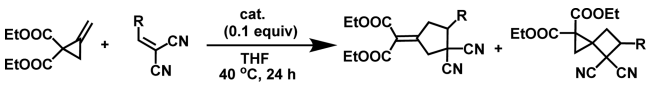
Table 1. Reaction of 1a with Alkenes 2 To Give Cyclopentylidenemalonates 3[a]



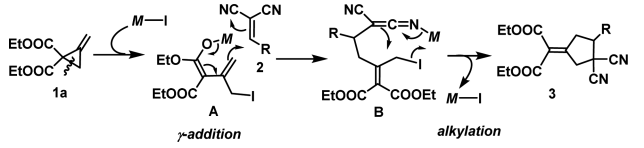
entry	2, R	cat.	3, yield (%)[b]	4, yield (%)
1	**2a**, Ph	–	**3a**, tr	**4a**, tr
2		Bu_2SnI_2	**3a**, tr	**4a**, tr
3		$MgBr_2$	**3a**, 47	**4a**, 34
4		MgI_2	**3a**, 83	**4a**, 12
5		Bu_2SnI_2–MgI_2	**3a**, 80	**4a**, 2
6	**2b**, *p*-ClC_6H_4	MgI_2	**3b**, 93	**4b**, tr
7		Bu_2SnI_2–MgI_2	**3a**, 78	**4b**, 4
8	**2c**, *p*-BrC_6H_4	MgI_2	**3c**, 68	**4c**, tr
9		Bu_2SnI_2–MgI_2	**3c**, 73	**4c**, tr
10	**2d**, *p*-FC_6H_4	MgI_2	**3d**, 61	**4d**, 3
11		Bu_2SnI_2–MgI_2	**3d**, 76	**4d**, 4
12	**2e**, *p*-$NO_2C_6H_4$	MgI_2	**3e**, 72	**4e**, tr
13		Bu_2SnI_2–MgI_2	**3e**, 90	**4e**, tr
14	**2f**, *p*-$MeOC_6H_4$	MgI_2	**3f**, 55	**4f**, 20
15		Bu_2SnI_2–MgI_2	**3f**, 57	**4f**, 20
16	**2g**, *p*-MeC_6H_4	MgI_2	**3g**, 61	**4g**, tr
17		Bu_2SnI_2–MgI_2	**3g**, 74	**4g**, 17
18	**2h**, Np	MgI_2	**3h**, 82	**4h**, 7
19		Bu_2SnI_2–MgI_2	**3h**, 67	**4h**, 8

[a]Conditions: **1a** (0.75 mmol), **2a** (0.5 mmol), cat. (0.05 mmol), THF (1 mL), 25 °C, 24 h. [b]tr = trace.

+ 3]-adduct derived from the addition of the α-carbon of tin dienolate **A**, similar to that of isocyanate (Scheme 1, eq 2).[3] However, there are no vinyl protons in the ^{1}H NMR spectrum, and two sets of methylene protons and a set of methine protons were observed. The byproduct **4a** proved to be a spirocarbocycle structure. The products, cyclopentylidenemalonate **3a** and spiro[2,3]hexane-1,1-dicarboxylate **4a**, were formally classified as a distal-bond-cleaved [3 + 2]-annulation product and a ring-untouched [2 + 2]-cycloaddition product, respectively.

For the synthesis of cyclopentylidenemalonate **3**, a plausible reaction path is shown in Scheme 3. Initially, the metal iodide

Scheme 3. Formation of Cyclopentylidenemalonates 3

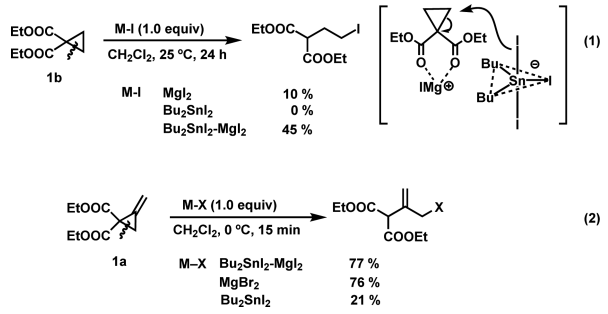


catalyst attacks **1a** at C3 to give metal dienolate **A**.[6] In a subsequent step, the dienolate adds to the alkene at the γ-carbon, giving metal keteneimine **B**. In the last stage, the intramolecular alkylation to alkyl iodide affords cyclopentylidenemalonate **3** and regenerates the catalyst. Thus, the reaction involves the catalytic employment of metal dienolate **A**.[7–9]

Table 1 also shows the results of the reaction of **1a** with various 1,1-dicyanoalkenes **2** under the optimized conditions (Table 1, entries 6–19). By the reactions with various 1,1-dicyanoalkenes **2** bearing functionalized aromatic substituents, the corresponding cyclopentylidenemalonates **3** were obtained with high selectivity. The levels of catalytic activity for both MgI_2 and Bu_2SnI_2–MgI_2 were comparable in all cases (Table 1, entries 6, 8, 10, 12, 14, 16, and 18 vs 7, 9, 11, 13, 15, 17, and 19).

We had already established that the characteristic feature of the tin halide ate complex $MgX^+[Bu_2SnBrX_2]^-$ is its hybrid makeup. MgX^+ acts as a Lewis acid to activate the dicarboxylate moiety to accelerate the ring opening of simple cyclopropanes (Scheme 4).[3] Simultaneously, a prolonged Sn–I bond that

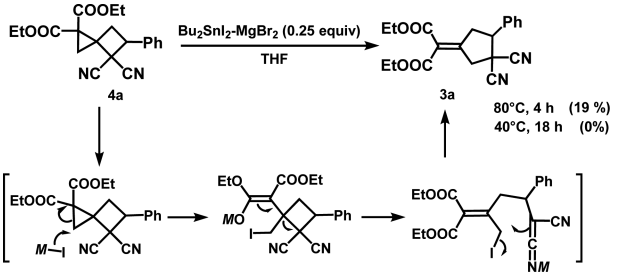
Scheme 4. Ring Opening of MCPs



occupies the axial position in the trigonal-bipyramidal tin structure[10] attacks the cyclopropane in an effective manner. When the tin complex was reacted with an equimolar amount of simple cyclopropane **1b** at rt for 24 h, a ring-opened adduct, iodoethyl malonate, was formed in 45% yield, whereas the use of only Bu_2SnI_2 or MgI_2 afforded lower yields (<10%) (Scheme 4, eq 1). In contrast, when an equimolar catalyst was reacted with methylenecycloproprane **1a** at 0 °C for 15 min, even the sole use of $MgBr_2$ was sufficient to initiate ring opening (Scheme 4, eq 2). As a result of its highly strained structure, methylenecycloproprane **1a** displayed a higher reactivity in the catalytic annulation when only a magnesium salt was used (Table 1, entries 3, 4, 6, 8, 10, 12, 14, 16, and 18).

There is a possibility of rearrangement from spirocarbocycle **4** to the stable adduct **3**. Thus, heating of **4a** in the presence of the catalyst afforded **3a**, albeit in a low yield (19%). The reaction proceeded via ring opening of the cyclopropane moiety, as shown in Scheme 5. However, when spirocarbocycle

Scheme 5. Rearrangement of 4a to 3a



4a was treated under the conditions shown in Table 1 (40 °C), no rearrangement occurred. Hence, cyclopentylidenemalonate **3a** was formed directly rather than by rearrangement of **4a**.

When dichloromethane instead of THF was used as the solvent, a dramatic change in the product (**3a**/**4a** = 11%/72%) was observed in the reaction of **1a** with **2a** catalyzed by Bu_2SnI_2–MgI_2 (Table 2, entry 1). Thus, the major product was **4a**.

Scheme 6 summarizes the mechanism for the syntheses of the two different types of products, **3a** and **4a**. Both products are derived from the same intermediate, keteneimine **B**,

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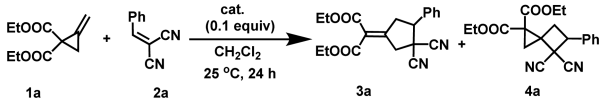
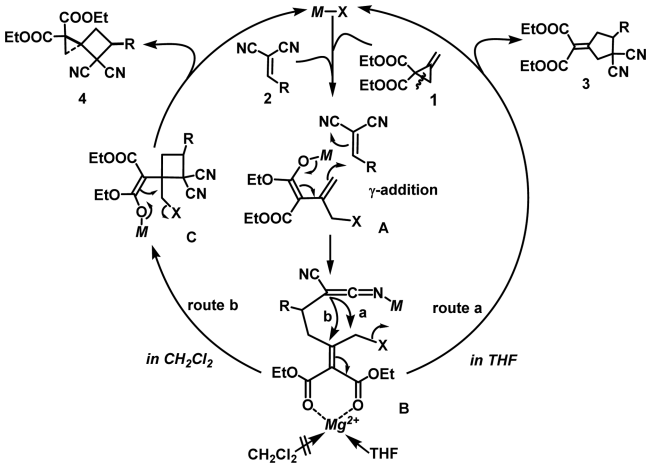
Table 2. Reaction of 1a with Alkene 2a To Give Spirocarbocycle 4a[a]



entry	cat.	3a, yield (%)[b]	4a, yield (%)
1	Bu_2SnI_2–MgI_2	11	72
2	MgI_2	28	58
3	$MgBr_2$	6	76
4	$MgCl_2$	tr	68
5	Bu_2SnBr_2–$MgBr_2$	tr	80
6	Bu_2SnCl_2–$MgCl_2$	tr	99

[a]Conditions: **1a** (0.75 mmol), **2a** (0.5 mmol), cat. (0.05 mmol), CH_2Cl_2 (1 mL), 25 °C, 24 h. [b]tr = trace.

Scheme 6. Catalytic Cycle for the Formation of 3 and 4



generated by the addition of the γ-carbon of dienolate **A**. The product is determined in the cyclization step. As solvents, the difference between dichloromethane and THF can be explained by their coordinating ability. THF, a coordinating solvent, aligns with Mg^{2+} and prevents the activation of the Michael acceptor moiety in **B**, and the intramolecular alkylation then proceeds to give **3** (route a). With dichloromethane, the effective activation of the Michael acceptor in **B** induces an intramolecular conjugate addition (route b). The resultant intermediate **C** causes a subsequent alkylation and gives spirocarbocycles **4**, which thereby regenerates the catalyst.

For the selective synthesis of spirocarbocycle **4a**, it is important to suppress the intramolecular alkylation (route a) in intermediate **B**. Accordingly, the halogen moiety in **B** was changed from an alkyl iodide either to alkyl bromide or chloride, both of which are less reactive than iodide. With the sole use of MgX_2, the reaction seemed to proceed by the same protocol as that with the tin ate complex. In fact, compared with MgI_2, using $MgBr_2$ and $MgCl_2$ catalysts afforded spirocarbocycle **4a** predominantly (Table 2, entries 2–4). It is noteworthy that the five-coordinate tin gave results that were superior to the sole use of MgX_2. Thus, when either the Bu_2SnBr_2–$MgBr_2$ or the Bu_2SnCl_2–$MgCl_2$ system was used as a catalyst, spirocarbocycle **4a** was obtained selectively, as expected (Table 2, entries 5 and 6). Among the catalytic systems examined, Bu_2SnCl_2–$MgCl_2$ was the best choice to afford the selective formation of **4a**. Thus, the five-coordinate tin was the preferred conjugate addition prior to alkylation.[11]

Synthetic and natural spirocarbocycles have attracted the attention of organic chemists because of their unique multidimensional structural features and numerous possible reactions whereby they undergo a carbon–carbon bond cleavage.[12] Among them, spiro[2,3]hexanes are rare structures. To date, there are few examples of the synthesis of spiro[2,3]hexanes wherein transition metal catalysts are necessary.[13,14] Table 3 summarizes the results of the reaction

Table 3. Reaction of 1a with Alkenes 2 To Give Spirocarbocycles 4[a]

entry	2, R	cat.	3, yield (%)[b]	4, yield (%)
1	**2a**, Ph	Bu_2SnCl_2–$MgCl_2$	**3a**, tr	**4a**, 99
2		$MgCl_2$	**3a**, tr	**4a**, 68
3	**2b**, *p*-ClC_6H_4	Bu_2SnCl_2–$MgCl_2$	**3b**, tr	**4b**, 95
4		$MgCl_2$	**3b**, tr	**4b**, tr
5	**2c**, *p*-BrC_6H_4	Bu_2SnCl_2–$MgCl_2$	**3c**, tr	**4c**, 98
6		$MgCl_2$	**3c**, tr	**4c**, 30
7	**2d**, *p*-FC_6H_4	Bu_2SnCl_2–$MgCl_2$	**3d**, tr	**4d**, 96
8		$MgCl_2$	**3d**, tr	**4d**, tr
9	**2e**, *p*-$NO_2C_6H_4$	Bu_2SnCl_2–$MgCl_2$	**3e**, tr	**4e**, 99
10		$MgCl_2$	**3e**, tr	**4e**, 40
11	**2f**, *p*-$MeOC_6H_4$	Bu_2SnBr_2–$MgBr_2$	**3f**, tr	**4f**, 55[c]
12		$MgBr_2$	**3f**, 31	**4f**, 33[c]
13	**2g**, *p*-MeC_6H_4	Bu_2SnCl_2–$MgCl_2$	**3g**, tr	**4g**, 96
14		$MgCl_2$	**3g**, tr	**4g**, 8
15	**2h**, Np	Bu_2SnCl_2–$MgCl_2$	**3h**, tr	**4h**, 97
16		$MgCl_2$	**3h**, tr	**4h**, 46

[a]Conditions: **1a** 0.75 mmol, **2** 0.5 mmol, cat. 0.05 mmol, CH_2Cl_2 1 mL, 25 °C, 24 h. [b]tr = trace. [c]The bromide catalyst gave a superior yield compared with the chloride catalyst.

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using various aromatic 1,1-dicyanoalkenes **2**. For the formation of **4**, Bu_2SnX_2–MgX_2 was a superior catalyst to MgX_2 alone in all cases (compare entries 1, 3, 5, 7, 9, 11, 13, and 15 vs entries 2, 4, 6, 8, 10, 12, 14, and 16). For the selective formation of **4**, three factors were important: the use of the noncoordinative solvent CH_2Cl_2, a metal chloride catalyst, and five-coordinate tin. Spiro[2,3]hexane-1,1-dicarboxylates **4** could be isolated in diastereomerically pure form, as shown by the ^1H and ^{13}C NMR spectra. For spirocarbocycle **4d** with a *p*-bromophenyl substituent, X-ray crystal analysis was successful.[15] The aryl and the carbon substituted for by dicarboxylates occupied the less-hindered *cis* positions in the wing-shaped cyclobutane ring.

In conclusion, we have developed a catalytic conversion of methylenecyclopropanes. The Mg–Sn catalytic system is an effective catalyst. Regioselectivity control is accomplished via the choice of a proper solvent and catalytic system. In particular, the synthesis of the rare spiro[2,3]hexane structure **4** is noted.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.orglett.7b01023.

Experimental procedures and spectral data (PDF)
X-ray data for compound **4c** (CIF)

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Notes

The authors declare no competing financial interest.

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