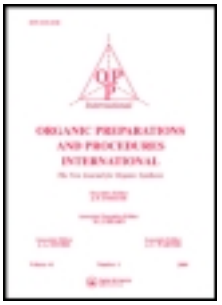

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REACTION OF METHACRYLOYL ISOCYANATE WITH SULFONIUM AND PHOSPHONIUM YLIDES

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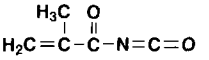
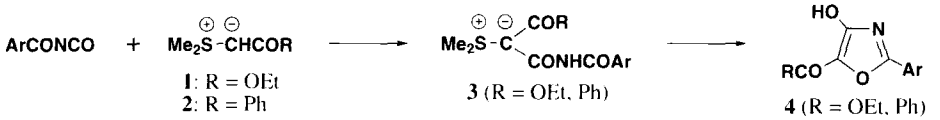
Methacryloyl isocyanate (**MAI**) is a versatile polyfunctional reagent bearing an enone moiety as well as a highly reactive acyl isocyanato group.[1-5] It is of particular interest that **MAI** functioned as an ambident heterodiene in cycloaddition reactions; enamines added not only to the acyl isocyanato moiety, but also to the enone moiety in **MAI**.[1]



MAI

It is known that alkyl and aryl isocyanates react with dimethylsulfonium (hereinafter abbreviated as sulfonium) ylides to afford the corresponding stable carbamoyl sulfonium ylides.[6,7] Similarly, benzoyl isocyanate easily reacted with carbonyl-stabilized sulfonium ylides such as **1** and **2**,[8] and triphenylphosphonium (hereinafter abbreviated as phosphonium) ylides[9] to give the corresponding benzoylcarbamoyl-stabilized sulfonium and phosphonium ylides, respectively. It has also been found that on thermolysis the above benzoylcarbamoyl-stabilized sulfonium ylides **3** are converted into oxazoles **4** (*Scheme 1*).[8]



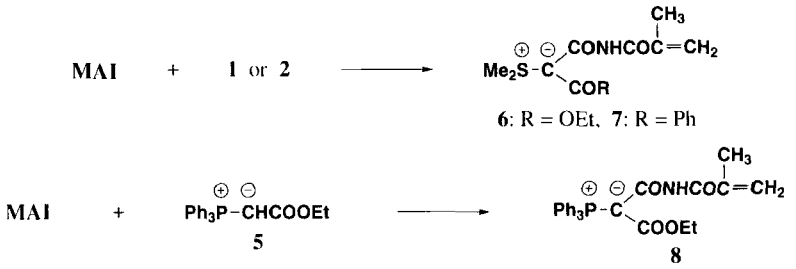
Scheme 1

It is also well known that carbonyl-stabilized sulfonium ylides react with α,β-unsaturated carbonyl compounds to furnish the corresponding cyclopropyl compounds through a Michael-type addition.[10-14] Only limited examples have been reported; however, carbonyl-stabilized phosphonium ylides are known to react with vinyl ketones to form the corresponding 1-acyl- (or 1-ethoxycarbonyl)-

4-oxoylides.[15]

The reaction of **MAI** with carbonyl-stabilized sulfonium and phosphonium ylides is particularly interesting, since **MAI** is an ambident reagent bearing acyl isocyanato and enone moieties. However, little attention has been paid to the reaction of **MAI** with these ylides. We wish now to report our findings on the reaction of **MAI** with carbonyl-stabilized sulfonium and phosphonium ylides, and thermolysis of the resulting carbamoyl ylides.

The carbonyl-stabilized ylides such as dimethylsulfonium ethoxycarbonylmethylide (**1**), dimethylsulfonium phenacylide (**2**) and triphenylphosphonium ethoxycarbonylmethylide (**5**) did not attack the enone moiety, but instead exclusively reacted with the isocyanato moiety in **MAI** at room tm temperature to afford the corresponding 1:1 adducts **6-8** in good yields (*Scheme 2*). The structures were assigned to the corresponding methacryloyl-carbamoylsulfonium ylides **6**, **7** and -phosphonium ylide **8** on the basis of their spectral data.



Scheme 2

As mentioned above, thermolysis of the carbamoyl-stabilized sulfonium ylides **3** gave oxazoles **4**.[8] It has also been reported that sulfonium ylide **2** is thermally stable but anhydrous copper(II) sulfate ($CuSO_4$) catalyzes its thermal decomposition.[11] Thus, thermolysis of carbamoyl-stabilized ylides **6-8** was investigated next.

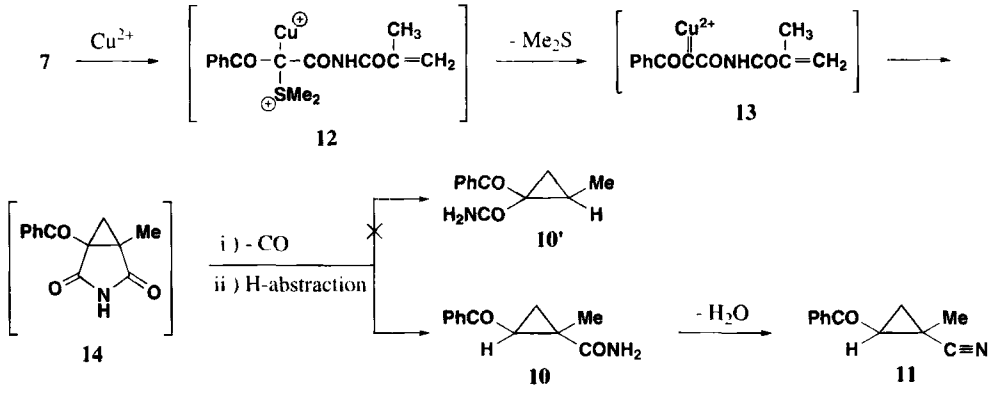
Although thermal decompositions were investigated under various conditions, ylides **6** and **8** bearing ethoxycarbonyl group yielded only intractable mixtures. On the other hand, **7** having a benzoyl group decomposed on heating in *m*-xylene or anisole under reflux to give a low yield of an unidentified product **9** whose molecular formula, $C_{26}H_{22}N_2O_6$, corresponded to that of a dimer of **7**, loss of dimethyl sulfide having occurred (*Entries 1 and 2* in Table 1). We have eventually found that refluxing a toluene or *m*-xylene solution of **7** with anhydrous $CuSO_4$ gave a mixture of two cyclopropyl compounds **10** and **11**. Since the yields of products were low, thermal decomposition products of **7**, obtained under various conditions, were investigated by use of gas-liquid chromatography (GLC) analyses, some of which are listed in Table 1 (*Entries 3-8*). It has been found that the thermolysis products obtained by use of a two-fold excess of anhydrous $CuSO_4$ in a dilute *m*-xylene solution, a moderate yield of a mixture of **10** and **11** was obtained (*Entry 6*). In thermolyses carried out on a preparative scale under the same conditions a mixture of **10** and **11** was obtained in a moderate yield (*Entry 10*).

On the basis of their spectral data as well as the consideration of the formation path illustrated in Scheme 3, the major product was assigned the structure of *r*-2-benzoyl-*t*-1-carbamoyl-1-methylcyclopropane (**10**) and the minor one the structure of *r*-2-benzoyl-*t*-1-cyano-1-methylcyclopropane (**11**).

Table 1. Thermolyses of Ylide **7**[a]

Entry	Amt. of 7 mmoles	Added salt	Molar ratio[b]	Solvent	Concen.[c] (mol/L)	Time (h)	Products (Yield/%)[d]
1	5.0	None		Anisole	0.6	5	**9** (12)
2	5.0	None		*m*-Xylene	0.6	2	**9** (18)
3	1.0	$CuSO_4$	1/0.5	*m*-Xylene	0.34	2	**10** (18), **11** (4)
4	1.0	$CuSO_4$	1/1	*m*-Xylene	0.34	2	**10** (20), **11** (4)
5	1.0	$CuSO_4$	1/2	*m*-Xylene	0.34	2	**10** (34), **11** (16)
6	1.0	$CuSO_4$	1/2	*m*-Xylene	0.07	2	**10** (44), **11** (8)
7	1.0	$Cu(acac)_2$[e]	1/2	*m*-Xylene	0.07	2	**10** (8), **11** (6)
8	1.0	$CuSO_4$	1/2	Toluene	0.07	2	**10** (28), **11** (8)
9	5.0	$CuSO_4$	1/2	*m*-Xylene	0.33	2	**10** (28), **11** (5)
10	4.0	$CuSO_4$	1/2	*m*-Xylene	0.07	2	**10** (39), **11** (6)

a) All the reactions were performed by refluxing solution of the ylide **7** in the stated solvent. b) Molar ratio of **7**/copper salt. c) Concentration of **7** in the stated solvent. d) Isolated yields in entries 1, 2, 9 and 10, and yields in entries 3-8 analyzed by gas-liquid chromatography (GLC) in entries 5-10 are shown. e) $Cu(acac)_2$: Copper(II) acetylacetonate.



Scheme 3

The reaction path illustrated in Scheme 3 may be conceivable for the formation of cyclopropanes **10** and **11**. In the copper-catalyzed thermolysis of sulfonium ylide **2**[11] or diazoacetophenone[16] a copper-complexed benzoylcarbene intermediate was suggested to be formed.

Accordingly, it is reasonable to consider the participation of a copper-complexed carbene intermediate in the copper-catalyzed thermal decomposition of ylide **7**. Thus, formation of a copper-carbon bond in **7** leads to **12**. Since Cu^{2+} has nine electrons available in its 3d orbital, $p\pi$-$d\pi$ overlap with simultaneous expulsion of dimethyl sulfide forms a copper-complexed carbene **13** bearing an enone moiety. The unique intramolecular cyclopropanation to the enone moiety in **13** gives the bicyclic intermediate **14**. Decarbonylation of **14**, followed by followed by hydrogen abstraction,[17] furnishes 1-benzoyl-2-carbamoyl-2-methylcyclopropane (**10**) in which benzoyl and carbamoyl groups are trans. Its regio isomer, 1-benzoyl-1-carbamoyl-2-methylcyclopropane (**10'**) was excluded on the basis of ^1H NMR data in which the cyclopropyl methylene proton appeared as a double doublet. Dehydration of **10** affords **11**. In fact, **10** furnished **11** when refluxed with anhydrous $CuSO_4$ in *m*-xylene.

EXPERIMENTAL SECTION

Melting points were determined on a Yanagimoto micro-apparatus and are uncorrected. IR spectra were measured as KBr pellets. ^1H and ^{13}C NMR spectra were recorded on a JEOL JNM-EX 270 in $CDCl_3$ and recorded at 270 and 67.5 MHz, respectively. Chemical shifts are expressed in parts per million downfield from Me_4Si. ^{13}C NMR resonance assignments were aided by the use of the DEPT technique to determine the number of attached hydrogen. Mass spectra were measured at 70 eV of ionization energy. The gas-liquid chromatography (GLC) analyses were carried out with a Yanagimoto GC-G2800 with flame detectors equipped with an OV-1 (5 mmϕ x 2 m) column using nitrogen as the carrier gas. The GLC yields were determined using nitrobenzene as an internal standard. Sulfonium ylide **1** is generated *in situ* from dimethyl (ethoxycarbonylmethyl)sulfonium bromide.[18] Sulfonium ylide **2**[18] and phosphonium ylide **5**[19] were prepared according to known procedures. **MAI** (bp. 121.5-122.5°/760 mmHg, 45°/45 mmHg) prepared by the reaction of methacrylamide with oxalyl chloride was supplied by the Nippon Paint Co. Ltd.(Osaka, Japan). Copper(II) sulfate and acetylacetonate were commercially available (Aldrich Chemical Company).

Reaction of MAI with Dimethylsulfonium Ethoxycarbonylmethylide (1).- A solution of ylide **1** was prepared by the following procedure. To a stirred suspension of dimethyl (ethoxycarbonylmethyl)sulfonium bromide (9.39 g, 41 mmol) in 35 mL of tetrahydrofuran (THF) was added sodium hydride (2.0 g, 50% dispersion in mineral oil, 41 mmol) in one portion. After the mixture had been stirred at room temperature for 12 h, it was filtered to remove the sodium bromide, thus giving a yellow solution of **1**.

After the solution of **1** was added to a solution of **MAI** (4.6 g, 41 mmol) in THF (1.5 mL), the mixture was stirred at room temperature for 12 h under nitrogen, during which time crystals were formed. Filtration gave 6.3 g of colorless crystals and the THF filtrate was evaporated *in vacuo* to leave a residue, which was washed with hexane (10 mL) to give 3.5 g of. colorless crystals. Recrystallization of the combined crystals (9.8 g, 92%) from benzene afforded pure dimethylsulfonium [1-(ethoxycarbonyl)-1-(methacryloylcarbamoyl)]methylide (**6**).

Compound 6: colorless needles, mp. 174-175° (dec.) IR: 3184, 1705, 1653, 1620, 1485, 1305, 1158, 1096 cm^{-1}; ^1H NMR: δ 1.29 (t, J = 7.2 Hz, 3H, ester-CH_3), 2.01 (d, J = 1.3 Hz, 3H, =CCH_3), 3.01 (s,

6H, SCH_3), 4.15 (q, J =7.2 Hz, 2H, CH_2), 5.50 (q, J = 1.3 Hz, 1H, =CH), 5.91 (br s, 1H, =CH), 12.03 (br s, 1H, NH); ^{13}C NMR: δ 14.63 (ester-CH_3), 18.44 (= CCH_3), 27.58 (SCH_3), 59.87 (CH_2), 61.56 (1-C), 121.02 (=CH_2), 140.97 (=C), 164.78, 166.32 (each N-C=O), 168.19 (ester-C=O); MS m/z (relative intensity): 259 (M^+, 2), 198 (10), 197 (M^+ - Me_2S, 4), 152 (197$^+$ - OC_2H_5, 5), 151 (4), 148 (M^+ - **MAI**, 1), 111 (**MAI**$^+$, 2)

Anal. Calcd for $C_{11}H_{17}NO_4S$: C, 50.96; H, 6.61; N, 5.40. Found: C, 51.03; H, 6.57; N, 5.54.

Reaction of MAI with Dimethylsulfonium Phenacylide (2).- A solution of ylide **2** (8.1 g, 45 mmol) in benzene (40 mL) was stirred with **MAI** (5.0 g, 45 mmol) at room temperature for 8 h under nitrogen, during which time colorless crystals were formed. Filtration and recrystallization from ethanol afforded 10.7 g (82%) of dimethylsulfonium (1-benzoyl-1-methacryloylcarbamoyl)methylide (**7**).

Compound 7: colorless prisms, mp. 177-178° (dec.). IR: 3010, 1702, 1657, 1630, 1543, 1489, 1162, 768, 706 cm^{-1}; ^1H NMR: δ 2.05 (d, J = 1.3 Hz, 3H, = CCH_3), 3.04 (s, 6H, SCH_3), 5.55 (q, J = 1.5 Hz, 1H, =CH), 6.06 (br s, 1H, =CH), 7.27-7.36 (m, 2H, ArH), 7.38-7.48 (m, 3H, ArH), 13.17 (br s, 1H, NH); ^{13}C NMR: δ 18.38 (CH_3), 27.92 (SCH_3), 79.17 (1-C), 121.99 (=CH_2), 126.47, 128.44, 129.68 (each ArC), 140.54 (=C), 141.45 (ArC), 164.49, 166.70 (each N-C=O), 189.74 (Ph-C=O); MS m/z (relative intensity): 291 (M^+, 1), 229 (M^+ - Me_2S, 6), 186 (M^+ - PhCO), 180 (M^+ - **MAI**, 10), 111 (**MAI**$^+$, 16), 105 (100).

Anal. Calcd for $C_{15}H_{17}NO_3S$: C, 61.83; H, 5.88; N, 4.81. Found: C, 62.02; H, 5.92; N, 4.81.

Reaction of MAI with Triphenylphosphonium Ethoxycarbonylmethylide (5).- A solution of ylide **5** (6.3 g, 18 mmol) in benzene (40 mL) was stirred with **MAI** (2.0 g, 18 mmol) at room temperature for 8 h under nitrogen, during which time crystals were formed. Filtration gave colorless crystals, which on recrystallization from benzene-hexane (1:1) afforded 7.5 g (90%) of triphenylphosphonium [1-(ethoxycarbonyl)-1-(methacryloylcarbamoyl)]methylide (**8**).

Compound 8: colorless needles, mp 171-172° (dec.). IR: 3214, 1709, 1655, 1618, 1481, 1441, 1299, 1158, 1110, 735, 690 cm^{-1}; ^1H NMR: δ 0.51 (t, J = 7.2 Hz, 3H, CH_3), 2.01 (d, J = 0.9 Hz, 3H, =C(CH_3)), 3.70 (q, J = 7.2 Hz, 2H, CH_2), 5.47 (q, J = 0.9 Hz, 1H, =CH), 5.95 (s, 1H, =CH), 7.40-7.59 (m, 9H, ArH) 7.61-7.75 (m, 6H, ArH), 12.18 (br s, 1H, NH); ^{13}C NMR: δ 13.46 (CH_3), 18.53 (=C(CH_3), 58.78 (CH_2), 59.24 (d, J_{CP} =122.1 Hz, P-C), 120.66 (=CH_2), 126.22 (d, J_{CP} = 94.0 Hz, ArC), 128.59 (d, J_{CP} = 12.3 Hz, ArC), 131.87 (d, J_{CP} = 3.7 Hz, ArC), 133.35 (d, J_{CP} = 9.7 Hz, ArC), 141.24 (=C), 165.96 (N-C=O), 166.75 (d, J_{CP} = 9.8 Hz, N-C=O), 170.12 (d, J_{CP} = 11.0 Hz, O-C=O); MS m/z (relative intensity) 459 (M^+, 3), 430 (M^+ - Et, 6), 279 (M^+- H_2C=C(CH_3)NH, 10), 278 (63), 277 (100), 263 (15), 262 (Ph_3P^+, 72), 261 (11) 201 (27), 197 (6), 185 (Ph_2P^+, 15), 184 (9), 183 (43),152 (13), 151 (11), 111(**MAI**$^+$, 20), 108 (20).

Anal. Calcd for $C_{27}H_{26}NO_4P$: C, 70.57; H, 5.71; N, 3.05. Found: C, 70.76; H, 5.69; N, 3.05.

Non-catalyzed Thermolysis of Carbamoyl-stabilized Ylide 7.- A solution of **7** (1.46 g, 5.0 mmol) in dry *m*-xylene (8 mL) was refluxed for 2 h, during which time 0.3 g (97%) of dimethyl sulfide was distilled off. After the reaction mixture had been concentrated *in vacuo*, the resulting yellow viscous

oily residue was triturated with benzene (3 mL) to give crystals which on recrystallization from ethyl acetate afforded 0.21 g (18%) of an unidentified dimer **9** as colorless needles, mp. 211-212° (dec.) (*Entry 2* in Table 1).

Compound 9: IR: 1771, 1688, 1584, 1251 cm^{-1}; ^1H NMR: δ 1.41 (d, J = 6.3 Hz, 6H, CH_3), 2.80-3.05 (m. 6H), 7.40-7.50 (m, 4H), 7.55-7.55 (m, 6H; ^{13}C NMR: δ 17.38 (CH_3), 32.38 (CH), 38.56 (CH_2), 91.86 (-C-), 128.37, 128.82, 133.83, 134.10 (each ArC), 184.90, 189.75, 197.10 (each C=O); MS *m/z* (relative intensity) 458 (M^+, 6) 353 (M^+ - PhCO, 3), 298 (3), 201 (2), 105 (100).

Anal. Calcd for $C_{26}H_{22}N_2O_6$: C, 68.11; H, 4.84; N, 6.11. Found: C, 67.85; H, 4.92; N, 6.09.

Copper-catalyzed Thermolysis of 7.- i) A typical procedure is given with an example of the isolation of products (*Entry 10* in Table 1). A solution of **7** (1.16 g, 4 mmol) in dry *m*-xylene (60 mL) was refluxed with anhydrous $CuSO_4$ (1.28 g, 8 mmol) for 2 h, during which time 151 mg (61%) of dimethyl sulfide was distilled off. Upon completion of the refluxing, any suspended solid (the copper salt) was removed by filtration, and the salt was washed thoroughly with the solvent. The filtrate was concentrated *in vacuo* and the residue was chromatographed (silica gel BW 300) to give 42 mg (6%) of *r*-2-benzoyl-*t*-1-cyano-1-methylcyclopropane (**11**) and 316 mg (39%) of *r*-2-benzoyl-*t*-1-carbamoyl-1-methylcyclopropane (**10**) from elution of benzene and benzene-ethyl acetate (1:1) respectively.

Compound 10: colorless prisms (cyclohexane), mp. 118-119°. IR: 3404, 3214, 1676, 1653, 1615 cm^{-1}; ^1H NMR: δ 1.33 (s, 3H, CH_3), 1.55-1.70 (m, 2H, CH_2), 3.36 (dd, J = 7.2, 7.6 Hz, 1H, CH), 6.05, 6.50 (each br s, 1H, NH), 7.4-7.6 (m, 3H, ArH), 7.95-8.0 (m, 2H, ArH); ^{13}C NMR: δ 13.19 (CH_3), 20.50 (1-C)), 29,96 (3-C),31.59 (2-C), 128.30, 128.66, 133.24, 137.95 (ArC), 175.67 (O=CNH_2), 196.71 (O=CPh); MS *m/z* (relative intensity) 203 (M^+, 0.2), 187 (M^+ - NH_2, 13), 186 (98), 159 (M^+ - $CONH_2$, 14), 158 (68), 105 (100).

Anal. Calcd for $C_{12}H_{13}NO_2$: C, 70.91; H, 6.45; N, 6.89. Found: C, 71.03; H, 6.57; N, 6.78.

Compound 11: colorless plates (hexane), mp. 55°. IR: 2242, 1680 cm^{-1}; ^1H NMR: δ 1.35 (s, 3H, CH_3), 1.69 (dd, J = 5.1, 8.4 Hz, 1H, 3-H), 1.78 (dd, J = 5.1, 6.7 Hz, 1H, 3-H), 3.25 (dd, J = 6.7, 8.4 Hz, 1H, 2-H), 7.45-7.70 (m, 3H, ArH), 7.95-8.10 (m, 2H, ArH); ^{13}C NMR: δ 14.57 (CH_3), 14.72 (1-C), 19.16 (3-C), 29.85 (2-C), 122.75 (CN), 128.34, 128.95, 133.89, 137.21 (ArC), 193.73 (C=O); MS *m/z* (relative intensity) 185 (M^+, 19), 158 (M^+ - HCN), 133 (5), 105 (100).

Anal. Calcd for $C_{12}H_{11}NO$: C, 77.81; H, 5.99; N, 7.56. Found: C, 77.72; H, 6.08; N, 7.41.

ii) The procedure for **entries 3-8** was as follows. After the thermolysis of **7** had been performed under the stated conditions, any suspended solid was filtered off and the solid was washed well with the solvent. The organic solution was concentrated *in vacuo* and the resulting residue was diluted with benzene (10 mL). To the benzene solution nitrobenzene was added as an internal standard and the mixture was analyzed by GLC. The components were identified by comparison of the retention times of pure **10** and **11**. The data in the table are given as the average of at least two parallel determinations.

Conversion of 10 to 11.- After a solution of **10** (203 mg, 1 mmol) in dry *m*-xylene (5 mL) had been refluxed with anhydrous $CuSO_4$ (800 mg, 5 mmol) for 10 h under argon, any suspended solid was

filtered off and the solid was washed with the solvent. The organic solution was concentrated *in vacuo* and the resulting residue was subjected to preparative TLC (benzene-ethyl acetate (3:1)) to provide 123 mg (61%) of unreacted **10** and 32 mg (17%) of **11**.

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