Mg-Promoted Reductive Cross-Coupling of Ethyl β-Arylacrylates with Aldehydes[1]

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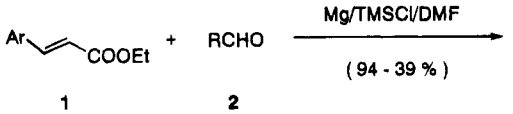
The skeleton of γ-lactones is very important and useful because it appears in many natural products including sesquiterpenes and alkaloids which sometime show interesting bioactivities, and it can be easily transformed to many useful skeletons such as pyrrolidines, butenolides, cyclopentanoids, and α-methylene-γ-butyrolactones.[2] Reductive cross-coupling of α,β-unsaturated esters with aldehydes seems one of the most useful and efficient methods to synthesize these γ-lactone skeletons, because the starting materials are in general easily prepared or commercially available. Only a few studies, such as an electroreductive method[3] and a SmI_2-promoted reaction in the presence of HMPA,[4] on this intermolecular coupling reaction have been reported. The synthetic utility of these methods may be, however, considerably limited because special equipment, carcinogenic solvent, and/or expensive reagents are required.

We now wish to report a facile method for selective carbon–carbon bond formation through Mg-promoted reductive cross-coupling of ethyl β-arylacrylates with aldehydes in the presence of trimethylchlorosilane (TMSCl) to give the corresponding γ-lactones in good to excellent yields (eq 1).



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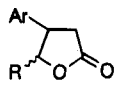


3 (1)

It was found that this reductive cross-coupling reaction was considerably influenced by the relative ratio of magnesium, TMSCl, and aldehydes **2** based on ethyl β-arylacrylates **1** as shown in Table 1, where the coupling of ethyl cinnamate (**1a**) with butyraldehyde (**2a**) was studied in detail as a typical example. It may be noteworthy that none of the cross-coupled product **3a** was obtained when TMSCl was absent in the reaction system while the presence of TMSCl let the cross-coupling reaction proceed smoothly to give 4-phenyl-5-propyldihydro-2(3*H*)-furanone (**3a**) in a good yield as an almost sole product. As shown on Table 1, the best result for formation of **3a** was obtained when the relative proportion of **1a**:Mg:TMSCl:**2a** was 1:3:3:15.

Table 1. Mg-Promoted Cross-Coupling of Ethyl Cinnamate (1a) with Butyraldehyde (2a) in the Presence of TMSCl

entry	Mg (mol equiv)	TMSCl (mol equiv)	**2a** (mol equiv)	yield of **3** (%)[a]
1	3	3	3	74
2	3	3	10	89
3	3	3	15	94
4	3	0	15	0
5	3	1.5	15	25
6	3	3	15	94
7	3	4	15	91
8	1.5	3	15	67
9	3	3	15	94
10	4	3	15	93

[a] Isolated yield.

Table 2. Mg-Promoted Cross-Coupling of α,β-Unsaturated Esters 1 with Aldehydes 2

			γ-lactones **3**	
entry	Ar in **1**	**2**	% yield[a]	(*cis*/*trans*)[b]
1	C_6H_5	CH_3CHO	**3a**	25 (1/1.4)
2	C_6H_5	paraldehyde	**3a**	91 (1/2)
3	C_6H_5	*n*-C_3H_7CHO	**3b**	94 (1/1.7)
4	C_6H_5	*iso*-C_3H_7CHO	**3c**	64 (1/1)
5	C_6H_5	*n*-C_4H_9CHO	**3d**	58 (1/1.7)
6	C_6H_5	*n*-$C_{11}H_{23}CHO$	**3e**	56 (–/1)
7	*p*-ClC_6H_4	*n*-C_3H_7CHO	**3f**	50 (1/1.5)
8	*p*-$CH_3OC_6H_4$	*n*-C_3H_7CHO	**3g**	49 (1/1.3)
9	*p*-$CH_3C_6H_4$	*n*-C_3H_7CHO	**3h**	39 (1/1.4)
10	α-$C_{10}H_7$	*n*-C_3H_7CHO	**3i**	68 (1/1.8)[c]
11	β-$C_{10}H_7$	*n*-C_3H_7CHO	**3j**	54 (1/2)[c]
12	3-thienyl	*n*-C_3H_7CHO	**3k**	39 (1/1.2)
13	2-thienyl	*n*-C_3H_7CHO	**3l**	47 (1/1.5)
14	3-piperonyl	*n*-C_3H_7CHO	**3m**	71 (1/1.3)

[a] Isolated yield. [b] Isomeric ratio determined by ^1H NMR analysis. [c] The reaction was carried out at 80 °C for 8 h.

Under the similar reaction conditions a variety of 4-aryl-5-alkyldihydro-2(3*H*)-furanones (**3**) were efficiently obtained in good to excellent yields through the present Mg-promoted reductive cross-coupling of the corresponding β-arylacrylates **1** with aliphatic aldehydes **2**, as shown in Table 2.[5]

The reductive cross-coupling reactions led to stereoisomeric mixtures of *cis* and *trans* γ-lactones. The ratio of *cis*/*trans* was determined by means of ^1H NMR and/or GC according to the assignments reported in the literature,[6] and in all of the cases, the *trans* isomers were obtained as the major products, which indicates importance of a steric effect of substituents in this reaction.

It was interesting that the present Mg-promoted method showed quite sharp contrast with hitherto known reactions for similar reductive cross-coupling of α,β-unsaturated esters with carbonyl compounds. Thus, this reaction did not give any cross-coupling product when

(1) Presented at the 67th National Meeting of Chemical Society of Japan, Tokyo, April 1994.

(2) (a) Nakamura, E.; Oshino, H.; Kuwajima, I. *J. Am. Chem. Soc.* **1986**, *108*, 3745. (b) Hoppe, D. *Angew. Chem. Int. Ed. Engl.* **1984**, *23*, 932. (c) Trost, B. M.; Bogdanowicz, M. J. *J. Am. Chem. Soc.* **1973**, 95, 5321. (d) Caine, D. D.; Frobese, A. S. *Tetrahedron Lett.* **1978**, 883. (e) Ehlinger, E.; Magnus, P. *Tetrahedron Lett.* **1980**, 21, 11. (f) Creger, P. L. *J. Org. Chem.* **1972**, *37*, 1907. (g) Eaton, P. E.; Cooper, G. E.; Johnson, R. C.; Muller, R. H. *J. Org. Chem.* **1972**, *37*, 1947.

(3) (a) Shono, T.; Ohmizu, H.; Kawakami, S.; Sugiyama, H. *Tetrahedron Lett.* **1980**, *21*, 5029. (b) Torii, S.; Inokuchi, T.; Hasegawa, N. *Chem. Lett.* **1980**, 639. (c) Froling, A. *Recl. Trav. Chem. Pays-Bas* **1974**, *93*, 47. (d) Sugino, K.; Nonaka, T. *Electrochim. Acta* **1968**, *13*, 613. (e) Kanetsuna, H.; Nonaka, T. *Denki Kagaku* **1979**, *47*, 422. (f) Nicolas, M.; Pallaud, R. *Comp. Rend. Soc. C* **1969**, *267*, 1834.

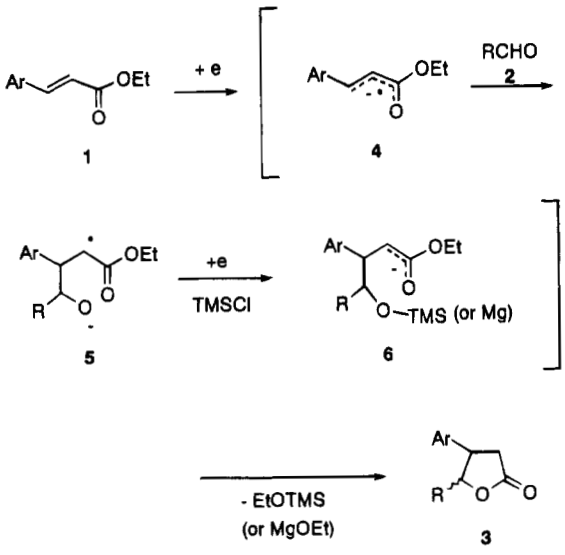
(4) (a) Otsubo, K.; Inanaga, J.; Yamaguchi, M. *Tetrahedron Lett.* **1986**, *27*, 5763. (b) Fukazawa, S.; Nakanishi, A.; Fujinami, T.; Sakai, S. *J. Chem. Soc. Chem. Commun.* **1986**, 624. (c) Girard, P.; Namy, J. L.; Kagan, H. B. *J. Am. Chem. Soc.* **1980**, *102*, 2693.

(5) All of the products **3a–m** were identified by spectroscopic methods such as ^1H and ^{13}C NMR, IR and mass spectra.

(6) (a) Ducher, S.; Michet, A. *Compt. Rend.* **1968**, *267C*, 1617. (b) Reissing, H.-U.; Angert, H.; Kunz, T.; Janowitz, A.; Handke, G.; Adjel, E. B. *J. Org. Chem.* **1993**, *58*, 6280.

0022-3263/95/1960-0458$09.00/0

Scheme 1



aliphatic acrylates were used as α,β-unsaturated esters.[7] On the other hand, in the reaction by the electroreductive or SmI_2-promoted method, use of aliphatic acrylates could bring about reductive cross-coupling with carbonyl compounds, and no cross-coupling product was obtained from the reaction of β-arylated-α,β-unsaturated esters with carbonyl compounds. Therefore, the present Mg-promoted reaction provides the first example for reductive cross-coupling of β-arylated-α,β-unsaturated esters with aldehydes giving the corresponding γ-butyrolactones.

Although the detail of the role of TMSCl in this coupling[8] still remains ambiguous, the reaction may be depicted as shown in the following scheme.

The reaction may be initiated through one-electron transfer from magnesium metal activated by TMSCl to β-arylacrylates **1** to give the corresponding anion radicals **4** (Scheme 1), which may be then subjected to electrophilic attack of aldehydes **2** activated by TMSCl, generating anionic radical species **5**, followed by the fast second electron transfer. Subsequently, the formed anionic cross-coupling intermediates **6** (possibly coordinated with Mg^{2+} ion or stabilized by TMSCl) may transformed to the product, γ-lactones **3**, through intramolecular cyclization.

Anyhow, the present method may be characterized by high simplicity and convenience of the reaction procedure, mild conditions (room temperature and ordinary pressure) and good yields of the products.

Experimental Section

^1H and ^{13}C NMR spectra were recorded at 270 and 75 MHz, respectively, using $CDCl_3$ as a solvent. TMS was used as an internal standard. IR spectra were obtained from neat films unless otherwise noted. MS were obtained at an ionization potential of 70 eV.

Ethyl β-arylacrylates except ethyl cinnamate were prepared by Wittig reaction of triphenylphosphonium ethyl acetate ylide and the corresponding aldehydes.[9] DMF was distilled from CaH_2 freshly prior to use. Commercially available Mg (turnings) was used as Mg metal. Other reagents such as aldehydes, TMSCl, and so on were obtained commercially and used without further purification.

General Procedure for Mg-Promoted Cross-Coupling of α,β-Unsaturated Esters and Aldehydes in the Presence of TMSCl. A solution containing ethyl β-arylacrylates (5 mmol), aldehydes (75 mmol), and TMSCl (15 mmol) in 50 mL of DMF was stirred under nitrogen atmosphere. Mg metal (15 mmol) was added to the solution in several portions which was then stirred for about 10 h at room temperature. After that, the reaction mixture was poured into 200 mL of saturated aqueous sodium bisulfite solution and was extracted by three 100-mL portions of ether. The combined ethereal solution was washed with three 100-mL portions of water and then dried over anhydrous magnesium sulfate. After removal of the drying agent by filtration, the solvent was evaporated by distillation. Then, the products, 4-aryl-5-alkyldihydro-2(3*H*)-furanones (**3**), were isolated by silica gel column chromatography of the resulting residue. Spectral data of the products showed that the obtained γ-lactones **3** were stereoisomeric mixtures of *cis* and *trans* isomers. The complete separation of *cis* and *trans* isomers by silica gel column chromatography was unsuccessful. The ratios of *cis*/*trans* are determined by means of ^1H NMR[6] and/or GC and summarized in Table 2. γ-Lactones **3a** and **3d** were identified by comparison of their ^1H and ^{13}C NMR with those of literature values.[6b]

4-Phenyl-5-propyldihydro-2(3*H*)-furanone (3b). ^1H NMR δ (ppm) *cis*: 0.81 (t, J = 7.29 Hz, 3H), 1.40 (m, 2H), 1.65 (m, 2H), 2.75 (m, 1H), 2.90 (m, 1H), 3.75 (m, 0.30H), 4.77 (m, 1H), 7.29 (m, 5H); *trans*: 0.89 (t, J = 7.56 Hz, 3H), 1.40 (m, 2H), 1.65 (m, 2H), 2.75 (m, 1H), 2.90 (m, 1H), 3.30 (m, 1H), 4.45 (m, 1H), 7.29 (m, 5H). ^{13}C NMR δ (ppm) *cis*: 13.71, 19.07, 33.61, 36.32, 45.90, 83.91, 126.77, 128.18, 137.99, 174.59; *trans*: 13.53, 19.36, 36.01, 36.17, 44.71, 86.75, 127.44, 127.67, 128.64, 139.75, 173.84; IR: ν 1780 cm^{-1}. MS: m/z 204 (M^+). Anal. Calcd for $C_{13}H_{16}O_2$: C, 76.44; H, 7.90. Found: C, 76.10; H, 7.95.

4-Phenyl-5-isopropyldihydro-2(3*H*)-furanone (3c). ^1H NMR δ (ppm) *cis*: 0.72 (d, J = 6.6 Hz, 6H), 1.90 (m, 1H), 2.66 (m, 1H), 3.04 (m, 1H), 3.63 (m, 1H), 4.34 (m, 1H), 7.25 (m, 5H); *trans*: 1.03 (d, J = 7.3 Hz, 6H), 1.68 (m, 1H), 2.66 (m, 1H), 3.04 (m, 1H), 3.43 (m, 1H), 4.25 (m, 1H), 7.25 (m, 5H). ^{13}C NMR δ (ppm) *cis*: 17.16, 18.70, 31.78, 38.13, 44.32, 89.89, 127.04, 127.92, 129.07, 140.77, 175.72; *trans*: 17.57, 19.80, 28.69, 38.65, 44.13, 91.42, 127.43, 127.92, 128.65, 139.09, 177.04. IR: ν 1780 cm^{-1}. MS: m/z 204 (M^+). Anal. Calcd for $C_{13}H_{16}O_2$: C, 76.44; H, 7.90. Found: C, 76.18; H, 7.48.

4-Phenyl-5-dodecyldihydro-2(3*H*)-furanone (3e). ^1H NMR δ (ppm) *trans*: 0.88 (t, J = 6.6 Hz, 3H), 1.23 (m, 16H), 1.47 (m, 2H), 1.67 (m, 2H), 2.74 (m, 1H), 2.95 (m, 1H), 3.30 (m, 1H), 4.44 (m, 1H), 7.30 (m, 5H). ^{13}C NMR δ (ppm) *trans*: 14–34 multiple, 37.48, 47.63, 86.97, 127–130 multiple, 139.06, 175.59; IR ν 1780 cm^{-1}. MS: m/z 316 (M^+). Anal. Calcd for $C_{21}H_{32}O_2$: C, 79.70; H, 10.19. Found: C, 79.51; H, 10.05.

4-(*p*-Chlorophenyl)-5-propyldihydro-2(3*H*)-furanone (3f). ^1H NMR δ (ppm) *cis*: 0.83 (t, J = 6.9 Hz, 3H), 1.43 (m, 2H), 1.68 (m, 2H), 2.71 (m, 1H), 2.96 (m, 1H), 3.70 (m, 1H), 4.71 (m, 1H), 7.24 (m, 4H). *trans*: 0.90 (t, J = 6.9 Hz, 3H), 1.43 (m, 2H), 1.68 (m, 2H), 2.71 (m, 1H), 2.96 (m, 1H), 3.29 (m, 1H), 4.40 (m, 1H), 7.24 (m, 4H). ^{13}C NMR δ (ppm) *cis*: 13.58, 19.10, 33.11, 37.32, 43.91, 83.62, 128.82, 129.07, 133.27, 136.67, 176.52; *trans*: 13.63, 18.90, 35.77, 36.01, 47.02, 86.48, 128.56, 129.14, 133.36, 137.53, 175.34. IR: ν 1780 cm^{-1}. MS: m/z 238 (M^+). Anal. Calcd for $C_{13}H_{15}ClO_2$: C, 65.41; H, 6.33. Found: C, 65.52; H, 6.11.

4-(*p*-Methoxyphenyl)-5-propyldihydro-2(3*H*)-furanone (3g). ^1H NMR δ (ppm) *cis*: 0.82 (t, J = 6.6 Hz, 3H), 1.38 (m, 2H), 1.68 (m, 2H), 2.70 (m, 1H), 2.92 (m, 1H), 3.69 (m, 1H), 3.80 (s, 3H), 4.69 (m, 1H), 7.04 (m, 4H); *trans*: 1.38 (m, 2H), 1.68 (m, 2H), 2.70 (m, 1H), 2.92 (m, 1H), 3.43 (m, 1H), 3.80 (s, 3H), 4.39 (m, 1H), 7.04 (m, 4H). ^{13}C NMR δ (ppm) *cis*: 13.62, 19.08, 31.00, 37.48, 43.66, 55.12, 84.04, 113.97, 128.69, 130.76, 158.93, 176.87; *trans*: 13.65, 18.95, 33.09, 35.88, 46.93, 55.05, 86.80, 114.33, 128.19, 130.76, 158.93, 175.63. IR ν 1780 cm^{-1}. MS: m/z 234 (M^+). Anal. Calcd for $C_{14}H_{18}O_3$: C, 71.77; H, 7.74. Found: C, 71.63; H, 7.52.

4-(*p*-Tolyl)-5-propyldihydro-2(3*H*)-furanone (3h). ^1H NMR δ (ppm) *cis*: 0.82 (t, J = 7.3 Hz, 3H), 1.42 (m, 2H), 1.68 (m, 2H), 2.34 (s, 3H), 2.70 (m, 1H), 2.92 (m, 1H), 3.60 (m, 1H), 4.70 (m,

(7) In these cases, the corresponding dimer of **1** or the hydrogenated β-aliphatic propionates were obtained as major products.

(8) In this reaction, three kinds of roles for TMSCl may be postulated; that is, activation of **2** as electrophile by coordination to the oxygen atom of the carbonyl group, stabilization of anionic intermediates generated by electron transfer from Mg metal, and activation of Mg metal.

(9) For example, Maercker, A. *Org. React.* **1965**, *14*, 270.

1H), 7.10 (m, 4H); *trans*: 0.89 (t, J = 7.3 Hz, 3H), 1.42 (m, 2H), 1.68 (m, 2H), 2.34 (s, 3H), 2.70 (m, 1H), 2.92 (m, 1H), 3.26 (m, 1H), 4.42 (m, 1H), 7.10 (m, 4H). ^{13}C NMR δ (ppm) *cis*: 13.54, 19.08, 35.74, 37.42, 44.06, 83.96, 127.54, 129.25, 135.00, 136.96, 176.94; *trans*: 13.62, 18.92, 33.06, 35.97, 47.29, 86.77, 127.00, 129.59, 135.86, 137.19, 175.70. IR: ν 1780 cm^{-1}. MS: m/z 218 (M^+). Anal. Calcd for $C_{14}H_{18}O_2$: C, 77.03; H, 8.31. Found: C, 77.15; H, 8.39.

4-(1-Naphthyl)-5-propyldihydro-2(3*H*)-furanone (3i). ^{1}H NMR δ (ppm) *cis*: 0.70 (t, J = 6.9 Hz, 3H), 1.43 (m, 2H), 1.74 (m, 2H), 2.88 (m, 1H), 3.14 (m, 1H), 4.16 (m, 1H), 5.05 (m, 1H), 7.70 (m, 7H); *trans*: 0.92 (t, J = 6.9 Hz, 3H), 1.43 (m, 2H), 1.74 (m, 2H), 2.88 (m, 1H), 3.14 (m, 1H), 3.50 (m, 1H), 4.78 (m, 1H), 7.70 (m, 7H). ^{13}C NMR δ (ppm) *cis*: 13.51, 18.99, 32.64, 37.30, 39.85, 82.42, 122–135 multiple, 175.14; *trans*: 13.80, 19.08, 33.22, 36.92, 41.92, 85.46, 122–135 multiple, 174.64. IR: ν 1780 cm^{-1}. MS: m/z 254 (M^+). Anal. Calcd for $C_{17}H_{18}O_2$: C, 80.28; H, 12.58. Found: C, 80.09; H, 12.65.

4-(2-Naphthyl)-5-propyldihydro-2(3*H*)-furanone (3j). ^{1}H NMR δ (ppm) *cis*: 0.79 (t, J = 7.3 Hz, 3H), 1.43 (m, 2H), 1.70 (m, 2H), 2.86 (m, 1H), 3.02 (m, 1H), 3.90 (m, 1H), 4.80 (m, 1H), 7.60 (m, 7H); *trans*: 0.90 (t, J = 7.3 Hz, 3H), 1.43 (m, 2H), 1.70 (m, 2H), 2.86 (m, 1H), 3.02 (m, 1H), 3.48 (m, 1H), 4.60 (m, 1H), 7.60 (m, 7H). ^{13}C NMR δ (ppm) *cis*: 13.72, 19.33, 33.36, 36.35, 44.77, 84.05, 122–135 multiple, 176.88; *trans*: 13.81, 19.10, 35.88, 37.57, 47.95, 86.71, 122–135 multiple, 175.70. IR: ν 1780 cm^{-1}. MS: m/z 254 (M^+). Anal. Calcd for $C_{17}H_{18}O_2$: C, 80.28; H, 12.58. Found: C, 80.36; H, 12.39.

4-(3-Thienyl)-5-propyldihydro-2(3*H*)-furanone (3k). ^{1}H NMR δ (ppm) *cis*: 0.86 (t, J = 6.9 Hz, 3H), 1.43 (m, 2H), 1.70 (m, 2H), 2.73 (m, 1H), 2.93 (m, 1H), 3.85 (m, 1H), 4.70 (m, 1H), 7.18 (m, 3H); *trans*: 0.94 (t, J = 6.9 Hz, 3H), 1.43 (m, 2H), 1.70 (m, 2H), 2.73 (m, 1H), 2.93 (m, 1H), 3.45 (m, 1H), 4.45 (m, 1H), 7.18 (m, 3H). ^{13}C NMR δ (ppm) *cis*: 14.14, 19.06, 36.20, 38.38, 41.43, 86.12, 122.11, 124.68, 128.04, 142.42, 171.94; *trans*: 13.83, 19.06, 37.19, 39.60, 42.92, 121.50, 126.01, 127.09, 140.66, 175.46. IR: ν 1780 cm^{-1}. MS: m/z 210 (M^+). Anal. Calcd for $C_{11}H_{14}O_2S$: C, 62.82; H, 6.71. Found: C, 62.65; H, 6.54.

4-(2-Thienyl)-5-propyldihydro-2(3*H*)-furanone (3l). ^{1}H NMR δ (ppm) *cis*: 0.85 (t, J = 6.9 Hz, 3H), 1.42 (m, 2H), 1.78 (m, 2H), 2.75 (m, 1H), 2.96 (m, 1H), 4.01 (m, 1H), 4.68 (m, 1H), 7.10 (m, 3H); *trans*: 0.92 (t, J = 7.3 Hz, 3H), 1.42 (m, 2H), 1.78 (m, 2H), 2.75 (m, 1H), 2.96 (m, 1H), 3.58 (m, 1H), 4.40 (m, 1H), 7.10 (m, 3H). ^{13}C NMR δ (ppm) *cis*: 13.74, 19.13, 32.86, 36.88, 40.19, 83.52, 124.47, 125.38, 127.13, 140.37, 175.80; *trans*: 13.78, 19.02, 35.90, 38.27, 42.89, 86.70, 124.47, 124.93, 127.31, 141.83, 174.80. IR: ν 1780 cm^{-1}. MS: m/z 210 (M^+). Anal. Calcd for $C_{11}H_{14}O_2S$: C, 62.82; H, 6.71. Found: C, 62.61; H, 6.56.

4-(3-Piperonyl)-5-propyldihydro-2(3*H*)-furanone (3m). ^{1}H NMR δ (ppm) *cis*: 0.83 (t, J = 7.3 Hz, 3H), 1.40 (m, 2H), 1.68 (m, 2H), 2.66 (m, 1H), 2.90 (m, 1H), 3.65 (m, 1H), 4.66 (m, 1H), 5.95 (s, 2H), 7.10 (m, 3H); *trans*: 0.89 (t, J = 7.3 Hz, 3H), 1.40 (m, 2H), 1.68 (m, 2H), 2.66 (m, 1H), 2.90 (m, 1H), 3.29 (m, 0.56H), 4.36 (m, 1H), 5.95 (s, 2H), 7.10 (m, 3H). ^{13}C NMR δ (ppm) *cis*: 13.80, 19.26, 33.13, 37.66, 44.36, 60.12, 84.05, 120.98, 131.85, 134.06, 146.98, 147.18, 148.31, 176.70; *trans*: 13.76, 19.04, 35.97, 36.09, 47.60, 60.37, 86.82, 120.64, 121.97, 132.73, 146.19, 147.11, 148.04, 175.54. IR: ν 1780 cm^{-1}. MS: m/z 248 (M^+). Anal. Calcd for $C_{14}H_{16}O_4$: C, 67.73; H, 6.50. Found: C, 67.59; H, 6.41.

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