Hydroacylation of Alkyl Vinyl Ketones and Acrylic Esters Using Organotetracarbonylferrates. Synthesis of 1,4-Dicarbonyl Compounds

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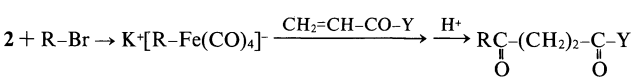
Alkyl vinyl ketones and acrylic esters were hydroacylated with organotetracarbonylferrates to the corresponding 1,4-diketones and 4-oxo carboxylic acid derivatives in good yield in dipolar aprotic solvents such as *N,N*-dimethylacetamide. Addition of 18-crown-6 improved the yield. *cis*-Jasmone and γ-jasmolactone were synthesized by use of these reactions.

Dicarbonyl compounds such as diketones, dicarboxylic acids, oxo carboxylic acids, and their derivatives are important intermediates in organic syntheses.[1] Many synthetic efforts have been conducted in view of this. However, syntheses of 1,4-dicarbonyl compounds are more difficult than their [1,3]direct condensation or [1,5]Michael addition counterparts, and this is reflected in the relative paucity of their synthetic routes.[2]

On the other hand, organotetracarbonylferrates (**1**) are known to react as acyl anion equivalents with various electrophiles to give carbonyl compounds including aldehydes, ketones, carboxylic esters, amides, and so on.[3] In this paper, we wish to report that **1**, prepared from potassium tetracarbonylferrate[4] and bromoalkanes, reacts with alkyl vinyl ketones and acrylic esters in dipolar aprotic solvents to give the dicarbonyl compounds in good yield.[5] A preliminary result has been described elsewhere.[6]

Results and Discussion

Synthesis of 1,4-Diketones and 4-Oxo Carboxylic Acid Derivatives by the Michael Addition. The results of this Michael addition type reaction of organotetracarbonylferrates with alkyl vinyl ketones and acrylic esters are listed in Table 1. For example, *cis*-8-undecene-2,5-dione was obtained in 55% yield starting from *cis*-1-bromo-3-hexene and potassium tetracarbonylferrate ($K_2Fe(CO)_4$, **2**) in *N,N*-dimethylformamide. Ethyl 4-oxodecanoate was obtained in 57% yield in *N*-methyl-2-pyrrolidone(NMP) (Scheme 1).

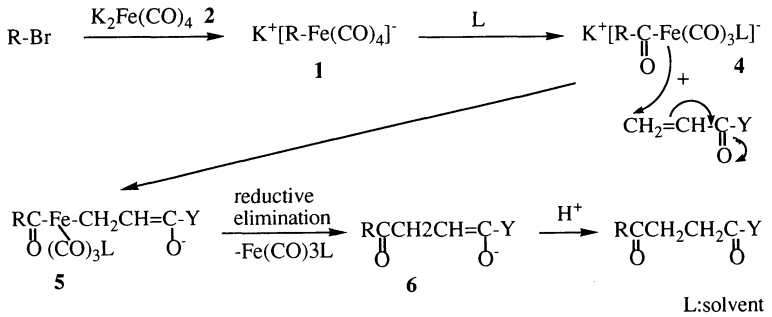


Scheme 1.

These reactions were dramatically affected by the nature of solvent used. The results of solvent change are also presented in Table 1, Runs 1—8, for the representative reaction of **1**, derived from 1-bromohexane and **2**, with methyl vinyl ketone. Thus, when *N,N*-dimethylacetamide was used as solvent, 2,5-undecanedione was obtained in 59% yield. When *N,N*-dimethylformamide was used, the yield was 53%. However, no diketone was obtained when tetrahydrofuran was used. Theses results indicate that a dipolar aprotic solvent such as *N,N*-dimethylacetamide is favorable for the reaction. The effects of these solvents may arise from an increase in the solubility of **1**, and also from an increase in the nucleophilicity by removal of the cation from a tight ion pairing situation in **1**.[7]

Next, the effect of the addition of 18-crown-6 **3** was investigated. The reaction of heptanoylcarbonylferrate with methyl vinyl ketone in the presence of **3** in NMP gave 2,5-undecanedione in 80% yield (Table 1, Run 5). Without **3**, the yield was 56% in NMP. A similar trend was noted also in the reaction of **1** with acrylic esters. Thus, addition of **3** increased the yields of diketones and keto esters. This effect of the cyclic ether may be due to complete removal of potassium cation from the ferrate **1**.

A possible reaction mechanism is illustrated in Scheme



Scheme 2.

Table 1. Hydroacylation of Alkyl Vinyl Ketones and Acrylates Using Organocarbonylferrates

Run	R–Br[a]	Michael acceptor	Solvent	Additive[b]	Product	Yield[c]/%
1	$CH_3CH_2CH_2CH_2CH_2CH_2$–Br	MVK[d]	DMAA[e]		$CH_3(CH_2)_5CO(CH_2)_2COCH_3$	70
2				C		84(70)
3			DMF[f]			64
4			NMP[g]			56
5				C		80(68)
6			HMPA[h]			56
7			DMSO[i]			40
8			THF[j]			0
9	$CH_3CH_2CH_2CH_2Br$	EVK[k]	DMAA		$CH_3(CH_2)_3CO(CH_2)_2COCH_2CH_3$	73
10			NMP			63
11	C_6H_5–$CH_2CH_2CH_2Br$	MVK	DMAA		C_6H_5–$(CH_2)_3CO(CH_2)_2COCH_3$	68
12				C		72
13	$(CH_3)_2CHCH_2Br$	EVK	DMAA		$(CH_3)_2CHCH_2CO(CH_2)_2COCH_2cH_3$	53
14	CH_3CH_2CH=$CHCH_2CH_2Br$ (cis)	MVK	DMF	C	CH_3CH_2CH=$CH(CH_2)_2CO(CH_2)_2COCH_3$	73(55)
15			DMAA			(52)
16		MA[l]	NMP		CH_3CH_2CH=$CH(CH_2)_2CO(CH_2)_2COOCH_3$	50(37)
17	$CH_3CH_2CH_2CH_2CH_2CH_2Br$	EA[m]			$CH_3(CH_2)_5CO(CH_2)_2COOCH_2CH_3$	57(40)
18	$CH_3CH_2CH_2CH_2CH_2CH_2CH_2Br$				$CH_3(CH_2)_6CO(CH_2)_2COOCH_2CH_3$	(36)
19	$CH_3CH_2CH_2CH_2CH_2CH_2CH_2CH_2Br$				$CH_3(CH_2)_7CO(CH_2)_2COOCH_2CH_3$	(26)
20	$CH_3CH_2CH_2CH_2CH_2Br$	MA			$CH_3(CH_2)_3CO(CH_2)_2cOOCH_3$	(27)
21	$CH_3CH_2CH_2CH_2CH_2CH_2Br$	EA			$CH_3(CH_2)_5CO(CH_2)_2COOCH_2CH_3$	47(57)
22		MA			$CH_3(CH_2)_5CO(CH_2)_2COOCH_3$	(28)
23		AN[n]			$CH_3(CH_2)_5CO(CH_2)_2CN$	(25)
24		MC[o]			$CH_3(CH_2)_5COCH(CH_3)CH_2COOCH_3$	(13)

a) Used for the preparation of organoferrate **1**. b) C denotes 18-crown-6. c) Determined by GLC. Yields in parentheses are isolated ones. d) Methyl vinyl ketone. e) *N*,*N*-dimethylacetamide. f) *N*,*N*-dimethylformamide. g) *N*-Methyl-2-pyrrolidone. h) Hexamethylphosphoric triamide. i) Dimethyl sulfoxide. j) Tetrahydrofuran. k) Ethyl vinyl ketone. l) Methyl acrylate. m) Ethyl acrylate. n) Acrylonitrile. o) Methyl crotonate.

2. An alkyl bromide first reacts with **2** to afford an alkylcarbonylferrate **1**, which is transformed into an acylcarbonylferrate **4** by solvent-assisted insertion of carbon monoxide. Reaction of **4** with an alkyl vinyl ketone or acrylic ester should lead to the acyl-aklyl iron complex **5** which appears to collapse by reductive eliminatin to the enolate ion **7** and an iron compound.[8] Protonation of the enolate anion **6** gives a 1,4-diketone or 4-oxo carboxylic ester.

Synthesis of 4-Oxo Carboxylic Esters by S_N2 Type Reaction. Besides the synthetic method described above, an alternative one for 4-oxo carboxylates was examined. In this reaction, heptanoyl- and *cis*-4-heptenoylcarbonylferrate were treated with methyl 3-bromopropionate in place of an acrylate (Scheme 3). The results are listed in Table 2.

$$Br(CH_2)_2COOCH_3 \xrightarrow{[RCOFe(CO)_3L]^-} RC(=O)(CH_2)_2C(=O)OCH_3$$

Scheme 3.

Although the corresponding 4-oxo carboxylates were obtained by this method, the yields were rather lower than those of the Michael addition method.

Synthesis of *cis*-Jasmone and γ-Jasmolactone. *cis*-Undecene-2,5-dione and methyl or ehtyl *cis*-4-oxo-7-decenoate are the synthetic precursors of the important perfume substances, *cis*-jasmone and γ-jasmolactone, respectively. Using the reaction described above, we have developed a convenient synthetic method of *cis*-jasmone and γ-jasmolactone in good overall yields.

cis-8-Undecene-2,5-dione was obtained from 1-bromo-*cis*-3-hexene in 55% yield. Base-catalyzed cyclization of the diketone gave *cis*-jasmone in 88% yield. *cis*-Jasmone was also prepared from 1-bromo-*cis*-3-hexene in 50% overall yield by one-pot reaction without isolation of the 1,4-diketone (Scheme 4).

γ-Jasmolactone was prepared from methyl or ehtyl *cis*-4-oxo-7-decenoate in good yield (70%) by reduction with $NaBH_4$ followed by treatment with 10 wt% NaOH aqueous solution and a phase transfer catalyst (benzyltriethylammonium chloride, BTEAC) in methanol (Scheme 5).

In conclusion, 1,4-diketones and 4-oxo carboxylic acid derivatives, the precursors of important perfume substances such as jasmones and jasmolactones, are obtained in moderate yield by the Michael addition type reaction of organocarbonylferrates **1** with alkyl vinyl ketones and acrylic esters.

Experimental

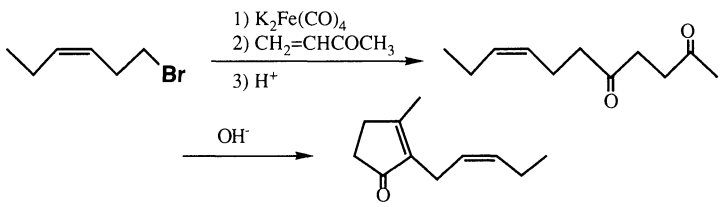
Infrared spectra (IR) were recorded on a Hitachi 260-10 Infrared Spectrophotometer as KBr pellets (for solids) or thin

Table 2. Alternative Synthesis of 4-Oxocarboxylic Esters and Jasmolactones

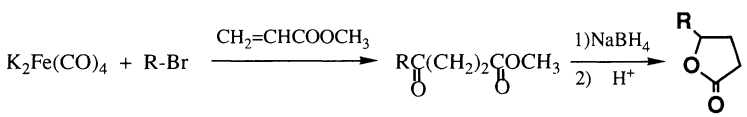
$$K_2Fe(CO)_4 + R\text{-}Br \xrightarrow{Br(CH_2)_2COOCH_3} \underset{\mathbf{A}}{RCO(CH_2)_2COOCH_3} \xrightarrow{1)\ NaBH_4\ \ 2)\ H^+} \underset{\mathbf{B}}{\text{R-γ-lactone}}$$

R	Yield of **A** (%)	Yield of **B** (%)[a]
C_6H_{13}–	40	68
$CH_3CH_2CH{=}CHCH_2CH_2$– (cis)	37	70

a) Isolated yield.



Scheme 4.



R: $CH_3CH_2CH{=}CHCH_2CH_2$- (*cis*)

Scheme 5.

films (for liquid) and the frequencies are given in reciprocal centimeters. ^{1}H NMR spectra were measured at 60 MHz in deuteriochloroform on a Hitachi R-600 FT-NMR Spectrometer and at 400MHz on a JEOL-400 Spectrometer using $CDCl_3$ as solvent, and all chemical shifts are expressed as valued in parts per million from internal tetramethylsilane. Coupling constants J are in Hz and splitting pattern abbreviations are s, singlet; d, doublet; t, triplet;, q, quartet; m, unresolved multiplet; br, broad. Analytical gas chromatography (GC) was performed on a Yanagimoto G-8 Model instrument using a stainless steel column packed with silicone gum SE-30 (10%) on chromosorb (0.3 cm×2 m) and a Shimadzu GC-14A instrument using a Shimadzu capillary column HiCAP CBP1-WR-100. Column chromatographies were performed on Merck silica gel 60 (230—400 mesh) and analytical thin layer chromatography was carried out on Merck 0.25 mm silica gel 60 plate F_{254}. Mass spectra were measured on a Hitachi RM-50 GC-MS instrument and recorded as m/z (rel intensity).

Material. All the reactions were conducted under an argon atmosphere. Tetrahydrofuran (THF) was dried and deoxygenated by distillation from potassium–benzophenone under an argon atmosphere just before use. Hexane was dried by distillation from sodium–benzophenone under argon just before use. Dipolar aprotic solvents, *N,N*-dimethylacetamide, *N,N*-dimethylformamide, *N*-methyl-2-pyrrolidone, hexamethylphosphoric triamide, and dimethyl sulfoxide were dried by distillation at reduced pressure from calcium hydride and stored over 3 Å molecular sieves under argon. Pentacarbonyliron (Strem Chemical Co.) was used without further purification. Potassium tri-*s*-butylhydroborate was purchased from Aldrich Chemical Co. as a 1.0 M (1 M= 1 mol dm^{-3}) THF solution under the trade name K-Selectride. Alkyl bromides, bromoethane, 1-bromopropane, 1-brompentane, 1-bromohexane, 1-bromohexadecane, 1-bromo-2-methylpropane, 1-bromo-3-methylbutane, and 1-bromo-3-phenylpropane were used without further purification. 1-Bromo-*cis*-3-hexene was prepared from *cis*-3-hexen-1-ol and phosphorus tribromide.[9] Methyl vinyl ketone, ethyl vinyl ketone, methyl acrylate, ethyl acrylate, and acrylonitrile were distilled under argon just before use. $K_2Fe(CO)_4$ was prepared according to the method described in the literature.[4] 18-Crown-6 and benzyltriethylammonium chloride (BTEAC) were commercial products of the highest purity and used without further purification.

General Procedure for the Preparation of 1,4-Dicarbonyl Compounds. An alkyl bromide (7 mmol) was added to a solution of $K_2Fe(CO)_4$ (10 mmol) in 30 ml of solvent and the reaction mixture was stirred for 1 h at room temperature before the addition of 5.8 ml of alkyl vinyl ketone or acrylate. After 20 h, 2 mol dm^{-3} hydrochloric acid was added to the reaction mixture. The solution was diluted with ether, washed three times with a saturated NaCl aqueous solution, dried ($MgSO_4$), filtered and concentrated. The residue was separated by column chromatography over silica gel (hexane–dichloromethane–diethyl ether 3 : 1 : 1).

The spectroscopic data of the representative compounds are as follows. Elemental analysis of these compounds gave satisfactory results for carbon and hydrogen.

2,5-Heptanedione. IR 2980, 1718, 1360, 1120 cm^{-1}. ^{1}H NMR δ=1.06 (t, 3H, J=7.2 Hz), 2.19 (s, 3H), 2.49 (q, 2H, J=7.2 Hz), 2.70 (s, 4H). MS (m/z) 128 (M$^+$).

2,5-Octanedione. IR 2960, 1710, 1365, 1162 cm^{-1}. ^{1}H NMR δ=0.92 (t, 3H, J=6.6 Hz), 1.45—2.10 (m, 2H), 2.18 (s, 3H), 2.44 (t, 2H, J=7.2 Hz), 2.69 (s, 4H). MS (m/z) 142 (M$^+$).

2,5-Decanedione. IR 2935, 1715, 1365, 1162 cm^{-1}. ^{1}H NMR δ=0.70—1.08 (m, 3H), 1.80—1.95 (m, 6H). 2.19 (s, 3H), 2.45 (t, 2H, J=6.6 Hz). MS(m/z) 170 (M$^+$).

2,5-Undecanedione. IR 2925, 1712, 1362, 730 cm^{-1}. ^{1}H NMR δ=0.88 (t, 3H, J=6.7 Hz), 1.27—1.85 (m, 8H), 2.19 (s, 3H), 2.45 (t, 2H, J=7.3 Hz), 2.69 (s, 4H). MS (m/z) 184 (M$^+$).

2,5-Heptadecanedione. IR 2920, 1700, 1418, 1362 cm^{-1}. ^{1}H NMR δ=0.85—1.90 (m, 23H), 2.19 (s, 3H), 2.20—2.70 (m, 2H), 2.69 (s, 4H). MS (m/z) 268 (M$^+$).

2,5-Henicosanedione. IR 2920, 1700, 1420, 1368 cm^{-1}. ^{1}H NMR δ=0.90 (t, 3H, J=5.4 Hz), 1.26—1.80 (m, 28H), 2.19 (s, 3H), 2.20—2.60 (m, 2H), 2.69 (s, 4H). MS (m/z) 324 (M$^+$).

7-Methyl-2,5-octanedione. IR 2970, 1718, 1370, 1168 cm^{-1}. ^{1}H NMR δ=0.87—0.97 (m, 7H), 2.19 (s, 3H), 2.30—2.52 (m, 2H), 2.69 (s, 4H). MS (m/z) 156 (M$^+$).

8-Methyl-3,6-nonanedione. IR 2950, 1710, 1365, 1164 cm^{-1}. ^{1}H NMR δ=0.88 (d, 6H, J=4.8 Hz), 1.14—1.80 (m, 3H), 2.19 (s, 3H), 2.45 (t, 2H, J=7.0 Hz), 2.69 (s, 4H). MS (m/z) 170 (M$^+$).

***cis*-8-Undecene-2,5-dione.** IR 2975, 1718, 1410, 1368 cm^{-1}. ^{1}H NMR δ=0.95 (t, 3H, J=7.0 Hz), 1.27—2.51 (m, 4H), 2.19 (s, 3H), 2.43 (t, 2H, J=4.0 Hz), 2.69 (s, 4H), 5.20—5.65 (m, 2H). MS (m/z) 182 (M$^+$).

8-Phenyl-2,5-octanedione. IR 2940, 1710, 1368, 1172, 750, 700 cm^{-1}. ^{1}H NMR δ=1.70—2.90 (m, 6H), 2.19 (s, 3H), 2.69 (s, 4H), 7.22 (m, 5H). MS (m/z) 218 (M$^+$).

Methyl *cis*-4-Oxo-7-decenoate. IR 2970, 1745, 1715, 1210, 735 cm^{-1}. ^{1}H NMR δ=0.96 (t, 3H, J=7.6 Hz), 1.73—2.80 (m, 10H), 3.63 (s, 3H), 5.37 (m, 2H). MS (m/z) 198 (M$^+$, 39), 167 (42), 115 (100), 55 (89).

Ethyl 4-Oxodecanoate. IR 2930, 1720, 1375, 1185 cm^{-1}. ^{1}H NMR δ=0.89 (t, 3H), 1.28 (m, 11H), 2.25—2.87 (m, 6H), 4.14 (q, 2H, J=6.8 Hz). MS (m/z) 214 (M$^+$, 2), 169 (25), 143 (100), 101 (53).

Ethyl 4-Oxoundecanoate. IR 2925, 1720, 1375, 1205 cm^{-1}. ^{1}H NMR δ=0.88 (t, 3H), 1.24 (m, 13H), 2.28—2.76 (m, 6H), 4.13 (q, 2H, J=6.8 Hz). MS (m/z) 228 (M$^+$, 4), 183 (48), 144 (94), 99 (100).

Ethyl 4-Oxotridecanoate. IR 2930, 1720, 1375, 1210 cm^{-1}. ^{1}H NMR δ=0.88 (t, 3H), 1.25 (m, 15H), 2.28—2.77 (m, 6H), 4.13 (q, 2H, J=6.8 Hz). MS (m/z) 242 (M$^+$, 2), 197 (22), 144 (97), 101 (100), 28 (90).

Methyl 4-Oxononanoate. IR 2945, 1735, 1435, 1360, 1210 cm^{-1}. ^{1}H NMR δ=0.88 (t, 3H), 1.32 (m, 6H), 2.10—2.77 (m, 6H), 3.66 (s, 3H). MS (m/z) 186 (M$^+$, 1), 155 (20), 130 (92), 115 (100). 99 (66).

Ethyl 7-Methyl-4-oxooctanoate. IR 2980, 1730, 1380, 1205 cm^{-1}. ^{1}H NMR δ=0.87 (d, 6H, J=5.2 Hz), 1.24 (t, 3H, J=7.2 Hz), 1.41—1.73 (m, 3H), 2.25—2.78 (m, 6H), 4.13 (q, 2H, J=7.2 Hz). MS (m/z) 200 (M$^+$, 1), 155 (45), 144 (76), 129 (31), 99 (100).

Methyl 4-Oxononanoate. IR 2935, 1730, 1370, 1210 cm^{-1}. ^{1}H NMR δ=0.88 (t, 3H), 1.28 (m, 8H), 2.19—2.93 (m, 6H), 3.67 (s, 3H). MS (m/z) 200 (M$^+$, 2), 169 (20), 130 (100), 115 (58), 98 (92).

4-Oxodecanenitrile. IR 2935, 2260, 1730, 735 cm^{-1}. ^{1}H NMR δ=0.90 (t, 3H), 1.28 (m, 8H), 2.30—2.87 (m, 6H). MS (m/z) 167 (M$^+$, 7), 113 (60), 71 (85), 43 (73).

Methyl 3-Methyl-4-oxodecanoate. IR 2925, 1740, 1200, 1180 cm^{-1}. ^{1}H NMR δ=0.89 (t, 3H), 1.04—1.53 (m, 11H),

2.21—3.28 (m, 5H), 3.64 (s, 3H). MS (*m*/*z*) 214 (M^+, 2), 182 (17), 143 (38), 113 (100), 85 (55), 43 (84).

Synthesis of *cis*-Jasmone. *cis*-8-Undecene-2,5-dione (0.124 g, 0.68 mmol) was added to a mixture of ethanol (3 ml) and 0.5 mol dm^{-3} aqueous NaOH (3 ml), and the mixture was refluxed for 4 h. The mixture was then diluted with water and extracted with ethyl acetate. The organic layer was washed with a saturated NaCl aqueous solution, dried over magnesium sulfate, filtered, and concentrated. Preparative TLC of the residual liquid gave *cis*-jasmone (0.103 g) in 92% yield.

***cis*-Jasmone.** IR 2960, 1698, 1642, 1381, 1069, 678 cm^{-1}. ^1H NMR δ=0.99 (t, 3H, J=7.6 Hz), 2.06 (s, 3H), 2.10—2.30 (m, 2H), 2.37 (t, 2H, J=4.6 Hz), 2.49 (d, 2H, J=4.9 Hz), 2.94 (d, 2H, J=6.7 Hz), 5.21—5.41 (m, 2H). MS (*m*/*z*) 164 (M^+).

Synthesis of γ-Jasmolactone.[10] Methyl *cis*-4-oxo-7-decenoate (2 mmol) in a 20 wt% NaOH aqueous solution (3 ml) and BTEAC (40 mg) were stirred at 50°C. After 2 h, $NaBH_4$ (3 mmol) in 2 ml of methanol was added. After stirring for 1 h at room temperature, the reaction mixture was acidified with 2 mol dm^{-3} H_2SO_4. After a usual workup, γ-jasmolactone was obtained as an oil in 70% yield.

γ-Jasmolactone. IR 2950, 1765, 1180 cm^{-1}. ^1H NMR δ=0.96 (t, 3H, J=7.2 Hz), 1.60—2.56 (m, 10H), 4.50 (m, 1H), 5.42 (m, 2H). MS (*m*/*z*) 168 (M^+, 4), 118 (9), 85 (28), 68 (100).

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