

Nilo Zanatta\*, Maria de F. M. Cortelini, Marcos J. S. Carpes, Helio G. Bonacorso, and Marcos A. P. Martins

Departamento de Química, Universidade Federal de Santa Maria (UFSM),  
97.119-900 - Santa Maria, RS, Brazil  
Received August 8, 1996

In this work the results of the reaction of  $\beta$ -alkoxyvinyl trihalomethyl ketones **1**, **2a-e**, with guanidine hydrochloride are reported. Depending on the ketone **1** or **2** and the conditions under which the reactions were carried out, 4-trihalomethyl-2-amino pyrimidines,  $\beta$ -alkoxyvinyl carboxylic acids, or  $\beta$ -acetal carboxylic esters were obtained.

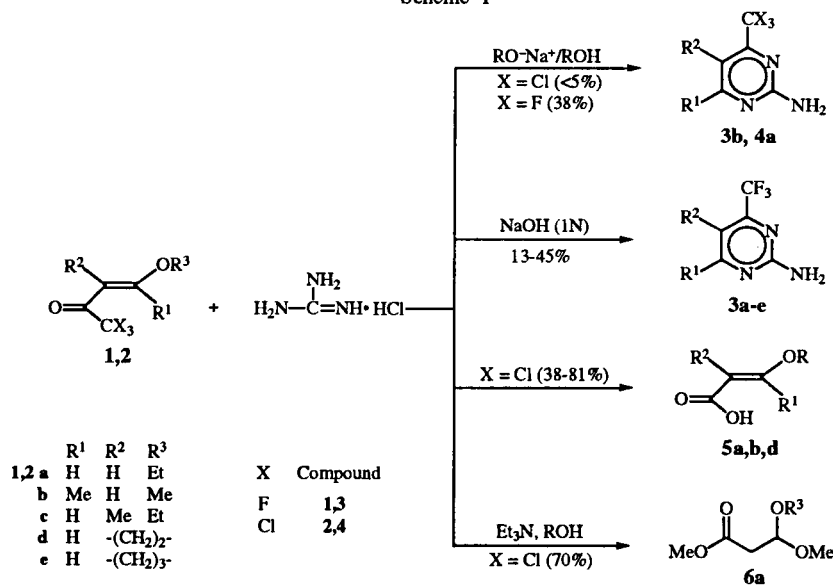
*J. Heterocyclic Chem.*, **34**, 509 (1997).

Aminopyrimidines and 2-aminopyrimidines substituted with simple groups, especially  $\text{CF}_3$ , are reported to possess various types of biological activities [1-4]. Due to the importance of 2-aminopyrimidines and other pyrimidines derivatives in the synthesis of highly effective drugs, numerous methods for their preparation have been reported [5,6]. 2-Aminopyrimidines which have a trifluoromethyl group at the 4-position have traditionally been synthesized from the reaction of the corresponding  $\beta$ -diketones [3],  $\beta$ -keto esters [7], and  $\beta$ -keto nitriles [8] with guanidine or any derivatives thereof. The reactions are usually carried out under alkaline conditions, *e.g.* sodium ethoxide/ethanol solution, but other solvents and neutral or acidic conditions have also been used. Recently the reaction of 1,1,1-trifluoro-4-ethoxy-3-buten-2-one with guanidine hydrochloride and guanidine carbonate was reported to give 2-amino-4-trifluoromethylpyrimidine in moderate yields [4].

As an extension of a series of cyclo-condensation reactions involving  $\beta$ -alkoxyvinyl trihalomethyl ketones **1**, **2** with a variety of nitrogen dinucleophiles of the type X-Y, such as hydroxylamine [9-12], hydrazine [13,14], and N-C-N such as urea *N*-methylurea [15,16], 2-methylthiourea [17], and amidines [18], developed in our laboratory, we now wish to report the synthesis of 4-trihalomethyl-2-aminopyrimidines **3a-e** and **4a**, from the reaction of  $\beta$ -alkoxyvinyl trihalomethyl ketones **1a-e** and **2a** with guanidine hydrochloride (Scheme 1). Depending on the conditions in which the reactions of the compound **2** (X = Cl) were carried out,  $\beta$ -alkoxyvinylcarboxylic acids **5** or  $\beta$ -acetalcarboxylic esters **6** were obtained, in good yields (see Scheme 1).

4-Trifluoromethyl-2-aminopyrimidines **3a-e** are easily prepared by the reaction of **1a-e** with guanidine hydrochloride in 1 *M* aqueous sodium hydroxide solution. A white precipitate appears as soon as the reactants are

Scheme 1



mixed under vigorous stirring at room temperature. Compounds **3a-e**, although, isolated in low to moderate yields (13-45%), no starting materials or side products were isolated suggesting that, in the presence of guanidine in 1 M sodium hydroxide solution, intensive decomposition of the starting  $\beta$ -alkoxyvinyl ketones to volatile or water soluble material indicating that retrograde aldol reaction had probably occurred [19]. Numerous unsuccessful attempts to improve the yields were made, however, the best results were obtained when aqueous sodium hydroxide solutions or alkoxides in alcohol solution were employed (see Scheme 1). The reason of the low yields is

probably because the "free" guanidine is a base strong enough to cause the decomposition of the  $\beta$ -alkoxyvinyl ketones **1** and **2**. A reaction carried out with the ketone **2b** and acetyl guanidine in methanol and reflux for 6 hours, the pyrimidine **3b** were obtained with 78% yield. This shows that by decreasing the basicity of the guanidine, the decomposition of the  $\beta$ -alkoxyvinyl ketones was mostly prevented and the yields of the cyclization was greatly improved. Selected physical and spectral data of compounds **3** and **4** are reported in Tables 1 and 2.

The synthesis of 4-trichloromethyl-2-aminopyrimidines from  $\beta$ -alkoxyvinyl trichloromethyl ketones **2a-e**

Table 1  
Selected Physical and Mass Spectral Data of **3a-e** and **4a**

No.	Yield (%) [a]	Mp [b] (°C)	Molecular Formula	Analysis (%) [c]			MS (70 eV) m/z [d]
				C	H	N	
<b>3a</b>	45	175	C <sub>5</sub> H <sub>4</sub> F <sub>3</sub> N <sub>3</sub> 163.10	36.82	2.47	25.76	163 (M+ 100), 144 (38), 136 (10), 94 (69), 67 (94)
				36.78	2.48	25.66	
<b>3b</b>	32	128	C <sub>6</sub> H <sub>6</sub> F <sub>3</sub> N <sub>3</sub> 177.13	40.69	3.41	23.72	177 (M+, 100), 158 (19) 130 (18), 108 (20), 81 (65), 69(22), 54 (23)
				40.75	3.58	23.78	
<b>3c</b>	44	150-154	C <sub>6</sub> H <sub>6</sub> F <sub>3</sub> N <sub>3</sub> 177.13	40.69	3.41	23.72	177 (M+, 92), 158 (31), 130 (18), 108 (15), 67 (100)
				40.53	3.43	23.48	
<b>3d</b>	13	159-160	C <sub>7</sub> H <sub>8</sub> F <sub>3</sub> N <sub>3</sub> O 207.16	40.59	3.89	20.28	208 (M+, 100), 190 (2), 176 (12)
				40.38	3.90	20.16	
<b>3e</b>	35	130-131	C <sub>8</sub> H <sub>10</sub> F <sub>3</sub> N <sub>3</sub> O 221.18	43.44	4.56	19.00	222 (M+, 82), 202 (44), 176 (100), 149 (15), 134 (15), 107 (7)
				43.35	4.71	18.89	
<b>4a</b>	<5	188	C <sub>5</sub> H <sub>4</sub> Cl <sub>3</sub> N <sub>3</sub> 212.47	28.27	1.90	19.78	213 (M+, 23), 176 (100), 149 (28), 141 (19), 115 (16), 94 (16), 67 (65), 52 (20)

[a] Yields of isolated compounds. [b] Melting points were determined on a Reichert Thermovar apparatus and are uncorrected. [c] Elemental analysis were performed on an Elementar Analysensysteme Vario EL apparatus. [d] The mass spectra were recorded on a Varian 3400 GC equipped with a Finnigan-MAT ITD 80A.

Table 2  
<sup>1</sup>H and <sup>13</sup>C NMR Data [a] of the Compounds **3a-e** and **4a**

Compound	<sup>1</sup> H nmr, $\delta$ , J (Hz)	<sup>13</sup> C nmr, $\delta$ , J <sub>C-F</sub> (Hz)
<b>3a</b>	6.8 (d, 1H, J = 4.9, H5), 7.2 (br s, 2H, NH <sub>2</sub> ), 8.5 (d, 1H, J = 4.9, H6)	163.6 (C-2), 154.7 (C-4, J = 34.6), 104.6 (C-5, J = 2.8), 161.4 (C-6), 120.6 (C-7, J = 275.4)
<b>3b</b>	2.3 (s, 3H, CH <sub>3</sub> ), 6.9 (s, 1H, H5), 7.1 (br s, 2H, NH <sub>2</sub> )	163.6 (C-2), 154.9 (C-4, J = 34.0), 104.4 (C-5, J = 2.8), 171.0 (C-6), 120.9 (C-7, J = 275.1), 23.7 (C-8)
<b>3c</b>	2.2 (q, 3H, J = 2.1, CH <sub>3</sub> ), 6.2 (br s, 2H, NH <sub>2</sub> ), 8.2 (s, 1H, H6)	160.7 (C-2), 152.0 (C-4, J = 30.2), 114.1 (C-5), 161.3 (C-6), 120.1 (C-7, J = 272.0), 12.4 (C-8)
<b>3d</b>	2.7 (t, 2H, J = 6.5, CH <sub>2</sub> ), 3.6 (t, 2H, J = 6.5, CH <sub>2</sub> ), 4.5 (br s, 1H, OH), 6.4 (br s, 2H, NH <sub>2</sub> ), 8.4 (s, 1H, H6)	160.5 (C-2), 150.0 (C-4, J = 28.9), 115.7 (C-5), 161.9 (C-6), 119.9 (C-7, J = 271.2), 28.7 (C-9), 59.9 (C-10)
<b>3e</b>	1.5 (t, 2H, J = 6.4, CH <sub>2</sub> ), 2.5 (t, 2H, J = 6.4, CH <sub>2</sub> ), 3.4 (t, 2H, J = 6.4, CH <sub>2</sub> ), 4.7 (br s, 1H, OH), 6.8 (br s, 2H, NH <sub>2</sub> ), 8.3 (s, 1H, H6)	162.2 (C-2), 152.2 (C-4, J = 32.9), 120.0 (C-5), 163.3 (C-6), 121.8 (C-7, J = 276.7), 34.3 (C-9), 24.4 (C-10), 60.4 (C-11)
<b>4a</b>	7.2 (d, 1H, J = 5.2, H5), 7.2 (br s, 2H, NH <sub>2</sub> ), 8.5 (d, 1H, J = 5.2, H6)	163.2 (C-2), 165.6 (C-4), 103.2 (C-5), 161.3 (C-6), 96.1 (CCl <sub>3</sub> )

[a] NMR spectra were recorded on a Bruker AC 80 (<sup>1</sup>H at 80 MHz and <sup>13</sup>C at 20 MHz) in DMSO-d<sub>6</sub>/TMS.

was mostly unsuccessful. Numerous attempts were made to extend the possible usefulness of these  $\beta$ -alkoxyvinyl trichloromethyl ketones to 2-aminopyrimidine synthesis. For example, attempts of condensing **2a-e** in sodium carbonate/ethanol, potassium hydroxide/ethanol, potassium hydroxide/water, Clay montmorillonite (K10), both acidic and basic aluminum oxide/methanol, and hydrochloric acid/methanol to cite but a few of the many conditions investigated, gave either very low yields (*e.g.* less than 5%), unchanged starting material, or non characterizable products.

During the tentative of the synthesis of 4-trichloromethyl-2-aminopyrimidines from the reaction of  $\beta$ -alkoxyvinyl trichloromethyl ketones **2a-e** with guanidine hydrochloride we found that these ketones were easily converted to the corresponding  $\beta$ -alkoxyvinyl carboxylic acids **5** when they were treated with 1 *M* solution of sodium hydroxide and in triethylamine/alcohol solutions, alkyl-3,3-dialkoxypropanoates **6** were obtained.

The reaction of  $\beta$ -alkoxyvinyl trichloromethyl ketones **2a,b,d** with guanidine hydrochloride in 1 *M* sodium hydroxide solution proceeded cleanly and without any side reaction to the corresponding  $\beta$ -alkoxyvinyl car-

boxylic acids **5a,b,d**, in good yields. Compound **2c** was not submitted to the reaction, and compound **2e** gave no isolated product (see Scheme 1).

Although, methods have been reported for the synthesis of  $\beta$ -alkoxy acids [20, 21], the yields in these procedures are not generally satisfactory. More recently, Hojo and co-workers [22] prepared **5a** in 90% yield, but the reaction conditions are more laborious.

Ketone **2a** when treated with guanidine hydrochloride in the presence of methanol and triethylamine, alkyl 3,3-dialkoxypropanoate **6a** was obtained, in good yields. Later we observed that the same reaction carried out without the guanidine hydrochloride also led to compound **6a**, in 20 minutes at 35°. Selected physical and spectral data of compounds **5** and **6** are reported in Tables 3 and 4.

Alkyl 3,3-dialkoxypropanoates are important intermediates for the synthesis of heterocyclic compounds such as coumarins [23], isoxazoles [24], porphyrins [25] and thiadiazines [26]. Due to the importance of compounds **6** numerous methods for their preparation have been published. These methods are multistep synthesis, affording mixtures of acetals and acrylates, requiring either expensive ethyl propiolate or giving poor overall yields [27-32].

Table 3  
Selected Physical and Mass Spectral Data of **5a,b,d** and **6a**

No.	Yield (%) [a]	Mp[bl (OC)	Molecular Formula	Analysis (%)			MS (70 eV) m/z [f]
				Calcd./Found [e]	C	H	
<b>5a</b>	36	110-112 [c]	C <sub>5</sub> H <sub>8</sub> O <sub>3</sub>	51.72	6.94	41.34	117 (M+1, 7), 98 (39), 87 (14), 69 (100)
			116.12	51.58	6.79	41.20	
<b>5b</b>	81	131	C <sub>5</sub> H <sub>8</sub> O <sub>3</sub>	51.72	6.94	41.34	116 (M+, 18), 99 (100), 87 (19), 69 (49), 59 (45), 55 (18)
			116.12	51.62	6.75	41.26	
<b>5d</b>	56	73-74 [c]	C <sub>5</sub> H <sub>6</sub> O <sub>3</sub>	52.63	5.30	42.07	114 (M+, 18), 96 (90), 68 (100), 55 (30), 53 (40)
			114.10	52.25	5.16	41.96	
<b>6a</b>	70	oil [d]	C <sub>7</sub> H <sub>14</sub> O <sub>4</sub>	51.84	8.70		[d]
			162.19	-	-		

[a] Yields of isolated compounds. [b] Melting points determined with a Reichert Thermovar apparatus and are uncorrected. [c] Known compound, see ref 19. [d] Known compound, see ref 33. [e] Elemental analysis were performed on a Elementar Analysensysteme Vario EL apparatus. [f] The mass spectra were recorded on a Varian 3400 GC equipped with a Finnigan-MAT ITD 80A.

Table 4  
<sup>1</sup>H and <sup>13</sup>C NMR Data [a] of the Compounds **5a,b,d** and **6a**

Compound	<sup>1</sup> H nmr, $\delta$ , J (Hz)	<sup>13</sup> C nmr, $\delta$ , J <sub>C-F</sub> (Hz)
<b>5a</b>	1.3 (t, 3H, J = 7.0, CH <sub>3</sub> ), 3.9 (q, 2H, J = 7.0, CH <sub>2</sub> ), 5.1 (d, 1H, J = 12.6), 7.6 (d, 1H, J = 12.6), 10.7 (br s, 1H, OH)	161.6 (C-1), 95.5 (C-2), 167.0 (C-3), 65.8 (C-4), 13.4 (C-5)
<b>5b</b>	2.3 (s, 3H, CH <sub>3</sub> ), 3.7 (s, 3H, OCH <sub>3</sub> ), 5.0 (s, 1H), 11.7 (br s, 1H, OH)	173.5 (C-1), 90.5 (C-2), 175.2 (C-3), 55.5 (C-4), 19.1 (C-6)
<b>5d</b>	2.7 (td, 2H, J = 9.2/0.5, CH <sub>2</sub> ), 4.5 (t, 2H, J = 9.2, CH <sub>2</sub> ), 7.3 (t, 1H, J = 0.5), 11.2 (br s, 1H, OH)	157.2 (C-1), 109.5 (C-2), 166.1 (C-3), 72.9 (C-4), 27.7 (C-7)
<b>6a</b>	1.2 (t, 3H, J = 7.1, CH <sub>3</sub> ), 2.7 (d, 2H, J = 5.9, CH <sub>2</sub> ), 3.4 (s, 3H, OCH <sub>3</sub> ), 3.6 (q, 2H, J = 7.1, CH <sub>2</sub> ), 3.7 (s, 3H, CH <sub>3</sub> ), 4.9 (t, 1H, J = 5.9, CH)	170.0 (C-1), 100.2 (C-2), 61.9 (CH <sub>2</sub> ), 53 (OCH <sub>3</sub> ), 51.3 (OCH <sub>3</sub> ), 40.0 (CH <sub>2</sub> ), 14.9 (CH <sub>3</sub> )

[a] Nmr spectra were recorded on a Bruker AC 80 (<sup>1</sup>H at 80 MHz and <sup>13</sup>C at 20 MHz) in CDCl<sub>3</sub>/TMS.

Recently, Tietze *et al* [33] reported the haloform reaction of 4-alkoxy-1,1,1-trichloro-3-buten-2-ones to give alkyl 3,3-dialkoxypropanoates in good yields. We describe here an efficient and simple method for the preparation of the compounds **6a**, which is easier and more rapid than that cited the literature [33].

#### Conclusion.

Although reaction of  $\beta$ -alkoxyvinyl trifluoromethyl ketones **1a-e** with guanidine hydrochloride furnished 4-trifluoromethyl-2-aminopyrimidines in low to moderate yields, the reaction of vinyl ketones (*e.g.* methyl vinyl ketone) with guanidine hydrochloride gives even lower yields. In turn, the reaction of  $\beta$ -alkoxyvinyl trichloromethyl ketones **2a-e** with guanidine hydrochloride in the presence of hydroxide or alkoxides undergo a fast haloform type reaction leading to the  $\beta$ -carboxylic acids **5** or alkyl 3,3-dialkoxypropanoates **6**.

#### EXPERIMENTAL

The  $\beta$ -alkoxyvinyl trihalomethyl ketones **1** and **2** were prepared according to reference [9].

General Procedure for the Synthesis of 4-Trifluoromethyl-2-aminopyrimidines **3a-e**.

##### Method A.

To a vigorously stirred solution of the  $\beta$ -alkoxyvinyl trifluoromethyl ketones **1a-e** (5.0 mmoles) and guanidine hydrochloride (7.5 mmoles) was added dropwise over 30 minutes, 6.0 ml of 1 *M* solution of sodium hydroxide. During the addition of the base a white precipitate was observed. The mixture was stirred for 10 minutes at room temperature. The precipitate was filtered, washed with distilled water, and dried overnight in a desiccator with silica gel under vacuum. All products were essentially pure according to capillary gc and  $^1\text{H}$  nmr analysis. Recrystallization from chloroform afforded the pure products **3a-e**.

##### Method B.

A solution of 0.57 g (6.0 mmoles) of guanidine hydrochloride in 5 ml of anhydrous ethanol was added to a solution of sodium ethoxide/ethanol obtained from 0.138 g (6.0 mmoles) of sodium in 10 ml of anhydrous ethanol. The resulting solution was filtrated to remove the sodium chloride. To the filtrated solution was added 0.84 g (5.0 mmoles) of **1b** and the solution was stirred under reflux for 4 hours. The solvent was evaporated to dryness and then 10 ml of distilled water was added. The resulting precipitate was removed by filtration, dried overnight in a desiccator with silica gel under vacuum, and recrystallized from chloroform.

General Procedure for the Synthesis of  $\beta$ -Alkoxyvinylcarboxylic Acids **5a,b,d**.

A 1 *M* aqueous solution of sodium hydroxide (5 ml, 7.5 mmoles) was added to the  $\beta$ -alkoxyvinyl trichloromethyl ketones **2a,b,d** (5.0 mmoles), dropwise over 0.5 hour. The mixture was stirred at room temperature for 2-2.5 hours and then cooled and acidified with 2 *M* hydrochloric acid until pH

~3. The resulting precipitate was removed by filtration and dried overnight in a desiccator with silica gel under vacuum. All the products were essentially pure by  $^1\text{H}$  nmr analysis. Recrystallization from chloroform afforded the pure products **5a,b,d**.

Methyl 3-Ethoxy-3-methoxypropanoate ester (**6a**).

A mixture of triethylamine (2 ml, 15.0 mmoles),  $\beta$ -alkoxyvinyl trichloromethyl ketone **2a** (10.0 mmoles) in 10 ml of dry alcohol (methanol or ethanol) was stirred at room temperature for 20 minutes. The mixture was evaporated under reduced pressure and then added 15 ml of a 0.1 *M* solution of hydrochloric acid. The solution was extracted with dichloromethane (2 x 15 ml) and the dichloromethane layer was washed with distilled water. The organic layer was dried with magnesium sulfate and evaporated to give **6a** which has been purified by column chromatography over silica gel with hexane/chloroform (9:1, v/v) as the mobile phase.

Acknowledgments.

The authors thank the financial support from the Conselho Nacional de Desenvolvimento Científico e Tecnológico (PADCT/CNPq), Fundação de Amparo à Pesquisa do Estado do Rio Grande do Sul (FAPERGS) and the fellowship from CNPq (M.F.M.C.) and (M.J.S.C.), are also acknowledged.

#### REFERENCES AND NOTES

- \* Author to whom correspondence should be addressed.
- [1] D. Lednicer and L. A. Mitscher, *Organic Chemistry of Drug Synthesis*, A Wiley-Interscience Publication, John Wiley & Sons, NY, V 1-3, 1977.
  - [2] A. G. Sorolla and A. Bendich, *J. Am. Chem. Soc.*, **80**, 5744 (1958).
  - [3] A. Kreutzberger, *J. Fluorine Chem.*, **29**, 385 (1985).
  - [4] I. I. Gerus, S. I. Vdovenko, M. G. Gorbunova, and V. P. Kukhar, *Chem. Heterocyclic Compounds*, 398 (1992).
  - [5] D. J. Brown, *The Pyrimidines*, Interscience, New York-London, 1962, p 206.
  - [6] D. J. Brown, *The Pyrimidines*, Suppl. 1, Wiley, New York-London, 1970, p 230.
  - [7] H. Gershon, A. T. Grefig, and A. A. Scala, *J. Heterocyclic Chem.*, **20**, 219 (1983).
  - [8] B. R. Baker and J. H. Jordaen, *J. Heterocyclic Chem.*, **2**, 162 (1965).
  - [9] A. Colla, M. A. P. Martins, G. Clar, S. Krimmer, and P. Fischer, *Synthesis*, 483 (1991).
  - [10] A. N. Zoch, G. Clar, N. Zanatta, H. G. Bonacorso, and M. A. P. Martins, *J. Heterocyclic Chem.*, **32**, 739 (1995).
  - [11] M. A. P. Martins, G. M. Siqueira, G. P. Bastos, and N. Zanatta, *J. Heterocyclic Chem.*, **33**, 1223 (1996).
  - [12] M. A. P. Martins, A. F. C. Flores, R. Freitag, and N. Zanatta, *J. Heterocyclic Chem.*, **33**, 1619 (1996).
  - [13] M. A. P. Martins, M. E. F. Braibante, G. Clar, *J. Heterocyclic Chem.*, **30**, 1159 (1993).
  - [14] M. A. P. Martins, R. Freitag, N. Zanatta, *Synthesis*, **12**, 1491 (1995).
  - [15] I. L. Pacholski, I. Blanco, N. Zanatta, and M. A. P. Martins, *J. Braz. Chem. Soc.*, **2**, 118 (1992).
  - [16] I. Blanco, I. L. Pacholski, N. Zanatta, and M. A. P. Martins, *Quím. Nova*, **16**, 15 (1993).
  - [17] C. C. Madruga, E. Clereci, M. A. P. Martins, and N. Zanatta, *J. Heterocyclic Chem.*, **32**, 735 (1995).

- [18] N. Zanatta, M. B. Fagundes, R. Hellenshon, H. G. Bonacorso, and M. A. P. Martins, *J. Heterocyclic Chem.*, Part 9, Submitted (1996).
- [19] J. March, *Advanced Organic Chemistry, Reactions, Mechanisms, and Structure*, John Wiley & Sons, New York, NY, 1992, 4th Ed, p 939.
- [20] G. Shaw and R. N. Warrener, *J. Chem. Soc.*, 153 (1958).
- [21] I. I. Kkolodkina, K. V. Levshina, S. I. Sergievskaya, and A. I. Kravchenko, *Zh. Org. Khim.*, 2, 66 (1966). Compound 5a was obtained by the hydrolysis of ethyl chlorocarbonylvinyl ether produced by reaction of  $\gamma$ -diethoxypropionic acid with thionyl chloride.
- [22] M. Hojo, R. Masuda, S. Sakaguchi, and M. Takagava, *Synthesis*, 25, 1016 (1986).
- [23] D. G. Crosby and R. V. Berthold, *J. Org. Chem.*, 27, 3083 (1962).
- [24] T. Kusumi, C. C. Chang, M. Wheeler, I. Kubo, and K. Nakamishi, *Tetrahedron Letters*, 22, 3451 (1981).
- [25] J. P. Collman, A. O. Chong, G. B. Jameson, R. T. Oakley, E. Rose, E. R. Schmittou, and J. A. Ibers, *J. Am. Chem. Soc.*, 103, 516 (1981).
- [26] P. Goya and M. Stud, *J. Heterocyclic Chem.*, 15, 253 (1978).
- [27] W. J. Croxall and H. J. Schneider, *J. Am. Chem. Soc.*, 71, 1257 (1949).
- [28] W. Truce and R. Heine, *J. Am. Chem. Soc.*, 79, 5311 (1957).
- [29] S. H. Berk, G. Dabbag, and P. Cotte, *J. Org. Chem.*, 47, 2216 (1982).
- [30] J. Walia and A. Walia, *J. Org. Chem.*, 41, 3765 (1976).
- [31] S. M. Elvain and R. L. Clarke, *J. Am. Chem. Soc.*, 69, 2657 (1947).
- [32] N. C. Deno, *J. Am. Chem. Soc.*, 69, 2233 (1947).
- [33] L. F. Tietze, H. Meier, E. Vob, *Synthesis*, 4, 274 (1988).