Synthesis of δ-cyclohexyl- and δ,δ-alkylene-α,α-dicarbonyl-substituted dienes and study of their valence isomerization

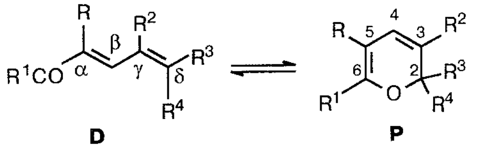
Zh. A. Krasnaya,[a]* ***V. S. Bogdanov,***[a] ***S. A. Burova,***[b] ***and Yu. V. Smirnova***[a]

[a]*N. D. Zelinsky Institute of Organic Chemistry, Russian Academy of Sciences,*
47 Leninsky prosp., 117913 Moscow, Russian Federation.
Fax: +7 (095) 135 5328
[b]*Higher Chemical College of Russian Academy of Sciences,*
9 Miusskaya pl., 125190 Moscow, Russian Federation.
Fax: +7 (095) 200 4204

β-Cyclohexylacrolein, β-cyclohexylmethacrolein, or α-cycloalkylidenalkanals were condensed with methyl acetoacetate or dimethyl malonate to give the δ-cyclohexyl- and δ,δ-alkylene-substituted α,α-dicarbonyl-containing α,β:γ,δ-dienes. The structures of the reaction products were studied using ^{1}H NMR, ^{13}C NMR, and UV spectroscopy. The diene keto esters bearing no substituents at the γ-position were shown to be in fact three-component equilibrium mixtures comprised of *E*- and *Z*-isomers of the diene (at the α,β bond) and a corresponding 2*H*-pyran. On the other hand, for keto esters with a Me group at the γ-position the equilibrium is shifted entirely to the 2*H*-pyrans. In contrast with the keto esters, dienic diesters exist only in the open form.

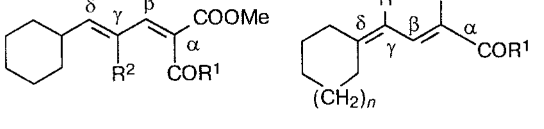
Key words: β-cyclohexylpropenals and α-cycloalkylidenalkanals; Knoevenagel condensation; α,β:γ,δ-dienones; 2*H*-pyrans; valence isomerization; effect of γ-substituents.

This work is devoted to further study of dienone (**D**) $\rightleftharpoons$ 2*H*-pyran (**P**) valence isomerization.



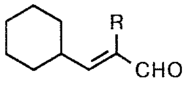
Earlier,[1—5] we discovered a considerable influence of substituents introduced into the dienone molecule, especially at the γ-position, on their ability for valence isomerization into corresponding 2*H*-pyrans, which allowed solvato-, thermo-, and photochromic properties to be exhibited by some dienones. The influence of δ-substituents remained poorly known owing to the absence of corresponding dienones.

δ-Cyclohexyl- and δ,δ-alkylene-α,α-dicarbonyl-containing dienes previously unknown were synthesized with the aim to study the influence of δ-substituents on valence isomerization:

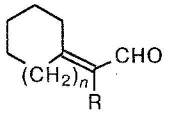


R^1 = Me, OMe; R^2 = H, Me; *n* = 0,1

Knoevenagel condensation between methyl acetoacetate (**1**) or malonate (**2**) and α,β-unsaturated aldehydes (**3**)—(**7**) was used to obtain these dienes:



3—4



5—7

3: R = H; **5,6**: *n* = 1; R = H (**5**); R = Me (**6**);
4: R = Me; **7**: *n* = 0, R = H

Aldehydes **3** and **4** were obtained *via* the oxypropenylation of cyclohexylmagnesium bromide with (trimethylsilyloxy)acroleins following an earlier described procedure.[6]

Aldehydes **5** and **6** were synthesized from 1,1-diethoxycyclohexane (**8**) *via* acetal-alkoxyalkene condensation through intermediates **9**—**12***.

The synthesis of compound **9** was described earlier[7]; however, we found that when using $FeCl_3$, recommended as the best catalyst in the work,[7] the yields of compound **9** were no more than 10%. The best yields of compounds **9** and **10** (76 and 61%, respectively) were obtained when $BF_3 \cdot OEt_2$ was used as catalyst.

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* A. A. Vasiliev is gratefully acknowledged for supplying aldehyde **7**, which he synthesized from cyclopentanone following the same scheme.

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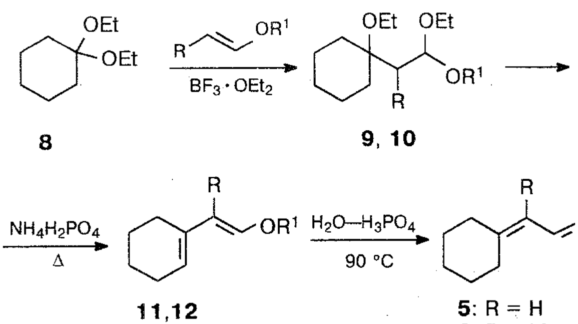


9,**11**: R = H, R^1= Et; **10**: R = R^1 = Me;
12: R = Me, R^1 = Et (**12a**), R^1 = Me (**12b**).*

The transformation of compound **10** to diene **12** was carried out in a 49% yield with the use of the procedure described in the work[8] for diene **11**, but at a higher temperature than for the latter. The hydrolysis of dienes **11** and **12** in the presence of phosphoric acid resulted in aldehydes **5** and **6** in 25 and 75% yields, respectively.

Condensation of aldehydes **3**—**7** with esters **1** and **2**, resulting in dicarbonyl-containing dienes (**13**—**21**), was carried out in the presence of piperidine or a mixture of piperidine and glacial AcOH, a part of the initial components being recovered unchanged. The reaction conditions and the substances thereby obtained which were isolated by vacuum distillation are given in Table 1; their structures were determined using UV spectra (Table 1) and ^{1}H and ^{13}C NMR (Tables 2 and 3). It follows from these data that diene ketoesters **13**, **17**, and **20**, obtained from ester **1** and aldehydes **3**, **5**, and **7**, are equilibrium mixtures of *E*- and *Z*-isomers at a α,β-double diene bond, **D**(*E*) and **D**(*Z*), and a corresponding 2*H*-pyran **P**.

Three sets of resonance signals of these compounds are observed in ^{13}C and ^{1}H NMR spectra. The signals were referred to the diene or 2*H*-pyran forms on the basis of a comparison of chemical shift and coupling constant values of compounds **13**, **17**, and **20** with that of dienones and 2*H*-pyran, studied earlier.[2,9] Referring the signals to *E*- and *Z*-isomers in ketoester **13** was made on the basis of *J* values of carbonyl carbon of acetyl group with proton H_β in both the isomers: $^3J_{CO,H\beta}$ = 9.5 Hz in *E*-isomer and $^3J_{CO,H\beta}$ = 6.2 Hz in *Z*-isomer, which correlates well with data obtained earlier.[10]

In accordance with our data concerning stereospecificity of chemical shifts for ^{13}C atoms of carbonyl groups in conjugated carbonyl-containing compounds[11] the signals were referred to *E*- and *Z*-isomers in ketoesters **17** and **20**: δ 200.77 and 200.67 in *E*-isomer and δ 195.17 and 195.30 in *Z*-isomer. It follows from the NMR data of *E*- and *Z*-isomers of ketoesters **13**, **17**, and **20** ($^3J_{\beta,\gamma}$ = 12—12.5 Hz) that *S-trans* conformation is realized there.

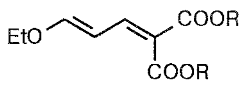
From the results of the study of temperature dependence of the isomeric composition of α,α-dicarbonyl-containing dienes investigated earlier,[2] it follows that compounds **13**, **17**, and **20** are also equilibrium mixtures of dienones **D**(*E*), **D**(*Z*) and 2*H*-pyrans (**P**).

Condensation products of ketoester **1** with aldehydes **4** and **6** are substituted 2*H*-pyrans **15** and **19**. There is one set of resonance signals referred to 2*H*-pyrans in ^{13}C and ^{1}H NMR spectra of compounds **15** and **19**. This referring is confirmed by UV spectra data as well (Table 1) because the most intense absorption band in the spectra of these compounds is found in a short-wave range: 207 nm (**15**) and 208 nm (**19**), which is consistent with UV spectra data of 2*H*-pyrans obtained earlier.[1]

Diene diesters **14**, **16**, **18**, and **21** were obtained by reaction of dimethyl malonate **2** with aldehydes **3**, **4**, **5**, and **7**.* According to ^{13}C and ^{1}H NMR they exist only in the open form and do not contain 2*H*-pyran.

Diesters **14**, **18**, and **21** exist in the form of *S-trans*-conformers ($^3J_{\beta,\gamma}$ = 12—12.5 Hz); diester **14** has a *trans*-position of protons at the γ,δ-bond ($^3J_{\gamma,\delta}$ = 15 Hz). There is one absorption maximum within the 274—294 nm range in UV spectra of diesters **14**, **16**, **18**, and **21**, which is characteristic of diene diesters.[1]

It should be noted that a pyran form is also not observed for either diene diester synthesized earlier, which contain a Me, Ph, or NMe_2 group at the δ-position.[3—5] We reported previously[12] that diene δ-ethoxy diesters (**22**; R = Me, Et) contained, according to NMR data, 20% of 2*H*-pyran (**23**), but this reference to the resonance signals of 2*H*-pyrans turned out to be wrong: they do not belong to 2*H*-pyrans, but to geometrical isomers of diene δ-ethoxy esters with *Z*-configuration of protons at γ,δ-double bond ($^3J_{\gamma,\delta}$ = 6 Hz).



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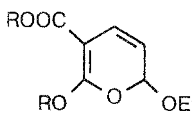

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The percentage of valence isomers in a $\mathbf{D}(E) \rightleftharpoons \mathbf{P}$ equilibrium mixture of keto esters **13**, **17**, and **20** in $CDCl_3$ is given in Table 1.** The data from this Table show that the most displacement of equilibrium to 2*H*-pyran is observed in keto ester **17**, that is probably connected with the destabilization of dienone due to steric hindrance caused by pentamethylene group.

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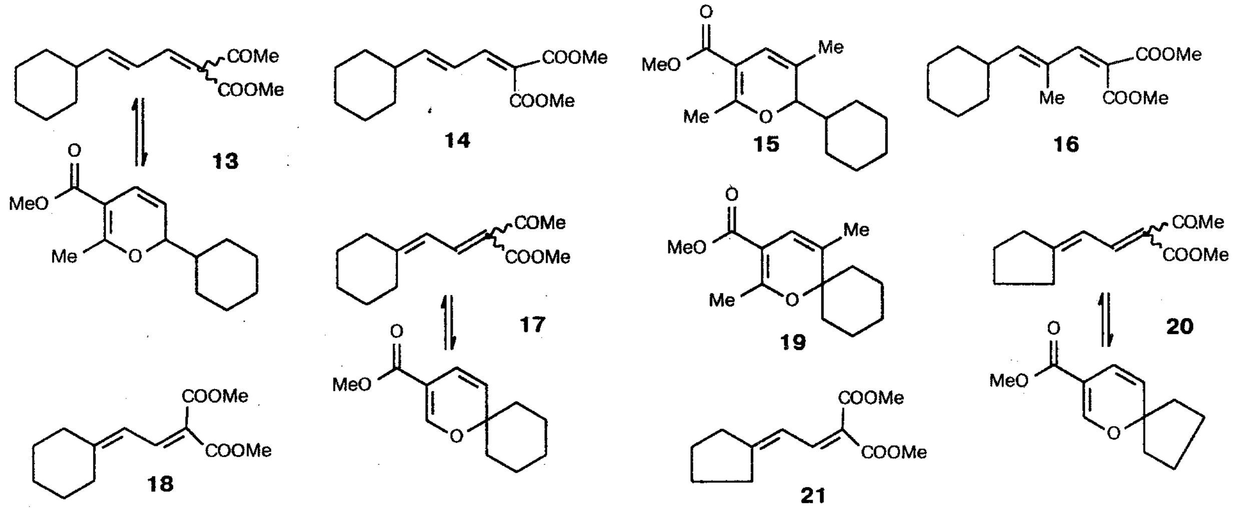
* Compound **12** seems to be a mixture of dienes with R^1 = Me and R^1 = Et.

* Attempts to obtain diester from compound **2** and aldehyde **6** failed.

** Content of valence isomers in CD_3OD is the same.

Table 1. Reaction conditions, yields, and constants of compounds **13—21**



Compound	Isomeric composition of the reaction product in the equilibrium (weight %)			$T_{reaction}$/°C (τ/days)	Yield* (%)	B.p./°C (*p*/Torr)	n_D^{20}	M$^+$, *m/z*	UV spectrum (EtOH) λ_{max}/nm (ε)
	E-**D**	**P**	*Z*-**D**						
13	29.7	23.3	47	10 (2)	60	108—112 (0.2)	1.5278	236	208(5200) 285(14800)
14	100	0	0	10 (1)	51	118—122 (0.2)	1.5245	252	278(24786)
15	0	100	0	10 (2)	25	102—105 (0.3)	1.5090	250	207(9545) 237(5682) 305(2727)
16	100	0	0	20 (30)	20	125—130 (0.2)	1.5120	266	274(26600)
17	15.8	67.2	17	10 (2)	20	97—105 (0.25)	1.5195	222	208(10185) 239(8095 306(6006)
18	100	0	0	20 (6)	20	153—155 (0.7)	1.5430	238	289(24911)
19	0	100	0	20 (4)	37	87—93 (0.25)	1.5100	236	208(16683) 238(11189) 300(6713)
20	46.8	31.2	22	10 (2)	16	83—87 (0.1)	1.5254		208(4784) 238(4160) 302(5616)
21	85	0	15	20 (9)	10	120—122 (0.3)	1.5279		294(25897)

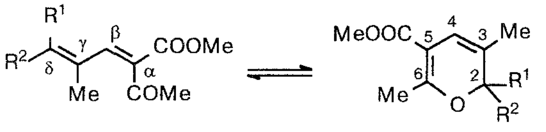
* Yield is calculated with account of the reagents taken for the reaction.

A particularly strong displacement of equilibrium, which makes it possible to observe only 2*H*-pyrans **15** and **19**, occurs when a proton in γ-position is substituted for a Me group. An analogous phenomenon was noted by us earlier.[1,4] The introduction of a Me group into γ-position of diene ketoesters **24** and **25** displaced the equilibrium completely to 2*H*-pyrans (**26**, **27**).

Table 2. ^{1}H NMR spectra of compounds **13—21***

Compound	δ (CDCl$_3$), *J*/Hz						
	Isomer	MeCO Me—C(6)—O	COOMe	Me at C$_\gamma$(3)	H$_\beta$(4)	H$_\gamma$(3)	H$_\delta$(2)
13	D(*E*)	2.37	3.76		7.25 d ($^3J_{\beta,\gamma}$ = 12)	6.2—6.56 m	
	D(*Z*)	2.32	3.83		7.21 d ($^3J_{\beta,\gamma}$ = 12)	6.2—6.56 m	
	P	2.25	3.68		6.2—6.56	5.2($^3J_{3,4}$ = 10; $^3J_{3,4}$ = 3.5)	4.44 m
14	D		3.75 3.82		7.35 ($^3J_{\beta,\gamma}$ = 12)	6.46 ($^3J_{\gamma,\delta}$ = 15; $^3J_{\beta,\gamma}$ = 12; $^4J_{\gamma,H'}$ = 1)	6.24 ($^3J_{\gamma,\delta}$ = 15; $^3J_{\delta,H'}$ = 7)
15	P	2.27	3.72	1.73	6.14		4.43 ($^3J_{H-2,H''}$ = 4.8)
16	D		3.76 3.81	1.73	7.24		5.82 ($^3J_{\delta,H'}$ = 7)
17	D(*E*)	2.40	3.80		7.68 ($^3J_{\beta,\gamma}$ = 12.5)	6.25 ($^3J_{\beta,\gamma}$ = 12.5)	
	D(*Z*)	2.34	3.86		7.65 ($^3J_{\beta,\gamma}$ = 12.5)	6.25 ($^3J_{\beta,\gamma}$ = 12.5)	
	P	2.30	3.72		6.32 ($^3J_{3,4}$= 10)	5.18 ($^3J_{3,4}$ = 10)	
18	D		3.77 3.83		7.74 ($^3J_{\beta,\gamma}$ = 12.5)	6.20 ($^3J_{\beta,\gamma}$ = 12.5)	
19	P	2.27	3.68	1.66	6.03		
20	D(*E*)	2.37	3.77		7.47 ($^3J_{\beta,\gamma}$ = 12.5)	6.43 ($^3J_{\beta,\gamma}$ = 12.5)	
	D(*Z*)	2.32	3.82		7.45 ($^3J_{\beta,\gamma}$ = 12.5)	6.43 ($^3J_{\beta,\gamma}$ = 12.5)	
	P	2.23	3.75		6.31 ($^3J_{3,4}$= 10)	5.16 ($^3J_{3,4}$ = 10)	
21	D		3.75 3.81		7.55 ($^3J_{\beta,\gamma}$ = 12.5)	6.38 ($^3J_{\beta,\gamma}$ = 12.5)	

* Ring protons: m, 1 — 2.43



24,25 **26,27**

24, 26: R^1 = R^2 = Me;

25, 27: R^1 = H, R^2 = Ph

Destabilizing influence of a bulky substituent at the γ-position on dienone is caused by a strong steric interaction between this substituent and one of substituents at C$_\alpha$, which takes place in an *S-trans*-conformer of dienone at its β,γ-bond.

Experimental

UV spectra were registered on a Specord UV-VIS spectrophotometer. ^{1}H NMR spectra were obtained with a Bruker WM-250 instrument (^{1}H 250 MHz) in relation to TMS; ^{13}C NMR spectra were obtained with a Bruker AM-300 instrument (75.432 MHz).

1-Ethoxy-1-(1'-methyl-2'-methoxy-2'-ethoxyethyl)cyclohexane (10). Propenyl methyl ether (15.4 mL, 0.21 mol) was added dropwise with stirring for 1 h to a mixture (cooled down to 0°C) of cyclohexanone diethylketal **8** (85.2 g, 0.5 mol) and 0.6 mL of BF$_3$.OEt$_2$. The reaction mixture was then stirred at 20°C for 1.5 h, neutralized with sodium methoxide, and fractionated *in vacuo*. On separating 67 g of ketal **8** that did not enter into the reaction, 16 g of β-alkoxyacetal **10** (31% taking account of propenyl methyl ether) were obtained, b.p. 140—160°C (7 Torr), n_D^{20} 1.4603.

1-Ethoxy-1-(2',2'-diethoxyethyl)cyclohexane (9). Following the above procedure, compound **9** was obtained from cyclohexanone and vinyl ethyl ether in 62% yield with account on vinyl ethyl ether.

β-(Cyclohexenyl-1)-propenyl methyl ether (12). A mixture of compound **10** (16 g, 0.07 mol) and NH$_4$H$_2$PO$_4$ (0.1 g) was heated in a Favorsky flask at 260—280 °C for 1 h. A distillate was obtained (b.p. 240—260 °C), washed with water, diluted with ether, and dried with potassium carbonate. On distillation, 4.6 g (49%) of compound **12** were obtained, b.p. 112—119 °C (7 Torr), n_D^{20} 1.4750.

Analogously, **β-(cyclohexenyl-1)-vinyl ethyl ether (11)** was obtained (yield 51%) from compound **9**.

2-Cyclohexylidenepropanal (6). A mixture of compound **12** (4.6 g, 0.03 mol) and 15% H$_3$PO$_4$ (30 mL) was stirred in a nitrogen stream at 90°C for 3 h; the reaction completion was determined with GLC. The reaction mixture was cooled down to 20°C, the upper layer was isolated, and the aqueous layer was extracted with ether. The combined organic layer was washed with a NaHCO$_3$ saturated solution and with water, and dried over MgSO$_4$. On distillation, 2.4 g (75%) of compound **6**

Table 3. ^{13}C NMR spectra of compounds **13—21**

Compound	Isomer	δ ($CDCl_3$) ($^1J_{^{13}C,^1H}$/Hz) [$^3J_{CO,H_\beta}$/Hz]*										
		$C_\alpha(5)$	$C_\beta(4)$	$C_\gamma(3)$	$C_\delta(2)$	MeCO		MeOCO		C(6)	$\underline{\mathrm{Me}}\mathrm{C}(6)$	$\underline{\mathrm{Me}}\mathrm{C}_\gamma(3)$
						Me	CO	MeO	CO			
13	**D**(*E*)	130.06	145.67 (156.5)	123.33 (156)	155.68 (156)	30.71	199.61 [9.5]	51.58	165.46 [7.1]			
	D(*Z*)	131.10	145.14 (155.5)	123.56 (158)	155.87 (156)	27.66	194.94 [6.2]	51.58	166.42 [12.1]			
	P	104.0	113.89 (165.4)	121.66 (166.4)	80.33 (146)			50.58	166.92	166.32	19.33	
14	**D**	122.65	146.65 (157)	123.21 (155)	155.34 (153)			52.03 51.97	165.57 164.99			
15	**P**	103.41	116.91 (164)	122.69	84.49 (149)			50.90	166.98	164.56	19.24	19.95
16	**D**	121.30	146.89 (156)	129.37	151.02 (152)			51.78 51.78	166.91 164.43			12.09
17	**D**(*E*)	129.57	139.30 (156)	118.06	161.21	29.80	200.77	51.72	166.54			
	D(*Z*)	130.64	138.95 (154)	117.70	160.42	28.34	195.17	51.72	167.17			
	P	103.23	120.06 (166)	119.96 (164)	78.75			50.7	166.55	165.83	20.94	
18	**D**	122.19	140.08 (155.5)	117.54 (156.5)	159.98			51.91 51.91	166.04 165.23			
19	**P**	104.14	116.24 (163)	128.19	80.37			50.55	166.58	163.36	19.13	17.57
20	**D**(*E*)	129.41	142.03 (154)	116.72	128.3	30.95	200.67	51.69	166.12			
	D(*Z*)	129.73	141.64 (154)	116.97 (167)	128.21	27.35	195.30	51.69	166.80			
	P	103.12	119.44 (163)	120.37 (167)	88.68			50.73	165.95	165.78	19.95	
21	**D**	120.80	142.50 (155.5)	116.37 (157.5)	165.07			51.71 51.71	165.77 165.25			

* Carbon rings: 41.15—42.26 (CH); 20.88—39.37 (CH_2).

were obtained, b.p. 90—99 °C (7 Torr), n_D^{20} 1.5025. UV spectrum (EtOH), λ_{max}/nm: 250 (ε 14081). ^{1}H NMR spectrum ($CDCl_3$, δ): 1.75 (s, 3 H, Me), 1.7—2.7 (m, 10 H, C_6H_{10}), 10.2 (s, 1 H, CHO).

Analogously, cyclohexylidenacetal **5** was obtained (yield 25%) from compound **11**, b.p. 75—80 °C (8 Torr), n_D^{20} 1.4965. UV spectrum (EtOH), λ_{max}/nm: 247. ^{1}H NMR spectrum ($CDCl_3$, δ, *J*/Hz): 1.7—2.7 (m, 10 H, C_6H_{10}), 5.81 (d, 1 H, CH), 10.0 (d, 1 H, CHO, *J* = 8 Hz).

Synthesis of compounds 13—15, 17, and 20. General procedure. Piperidine (0.1 mL) was added to a mixture (cooled down to −10°C) of aldehyde **3** (2 g, 0.0114 mol) and methyl acetoacetate **1** (1.7 g, 0.0114 mol), and the reaction mixture was allowed to stand at 10°C for 2 days. Then 20 mL of ether and 10 mL of benzene were added and the mixture was treated with water. On separating aqueous layer, the organic layer was washed with 1% HCl and with water, dried over $MgSO_4$, and distilled. After compounds **3** and **1** which did not enter into the reaction were removed, 2 g of compound **13** (60%) were obtained, b.p. 108—112 °C (0.2 Torr). Found (%): C 71.46; H, 8.42. $C_{14}H_{20}O_3$. Calculated (%): C, 71.16; H, 8.53.

Analogously, diester **14** was obtained from dimethyl malonate **2** and aldehyde **3**. Found (%): C, 66.44; H, 7.64. $C_{14}H_{20}O_4$. Calculated (%): C, 66.64; H, 7.99.

Reaction conditions and the constants of compounds **14**, **15**, **17**, and **20** are given in Table 1.

Synthesis of compounds 16, 18, 19, and 21. General procedure. A mixture of dry piperidine (0.1 mL), glacial AcOH (0.05 mL), and of benzene (0.5 mL) was added to a mixture of aldehyde **5** (1 g, 0.008 mol) and dimethyl malonate **2** (1 g, 0.008 mol). After the reaction mixture was allowed to stand at 20°C for 6 days, it was diluted with ether (15 mL) and benzene (10 mL) and then washed with 3% H_2SO_4 and with water. The organic layer was dried over $MgSO_4$ and evaporated, the rest was fractionated *in vacuo*. On removing compounds **5** and **2** which did not enter into the reaction, 0.4 g (20%) of compound **18** was obtained, b.p. 153—155 °C (0.7 Torr). Found (%): C, 65.35; H, 7.48. $C_{13}H_{18}O_3$. Calculated (%): C, 65.53; H, 7.61.

Analogously, 2*H*-pyran **19** was obtained from methyl acetoacetate **1** and aldehyde **6**. Found (%): C, 70.95; H, 8.38. $C_{14}H_{20}O_3$. Calculated (%): C, 71.16; H, 8.53.

Reaction conditions and the constants of diene diesters **16**, **18**, **21**, and pyran **19** are given in Table 1.

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