Michael additions of 1,3-cycloalkanediones to dimethyl acetylenedicarboxylate

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Michael additions of 1,3-cyclopentanedione, 1,3-cyclohexanedione, 5,5-dimethyl-1,3-cyclohexanedione, and 1,3-cycloheptanedione to dimethyl acetylenedicarboxylate catalyzed by CH_3ONa, piperidine or KF/Al_2O_3 were performed. There was found out that, like additions to a double bond, the Michael addition to a triple bond takes place successfully under KF/Al_2O_3 catalysis. The influence of a catalyst upon *cis*/*trans* ratio of the addition product was investigated; catalysis by KF/Al_2O_3 prefers the *cis* addition to the greater extent compared to catalysis by sodium methoxide. It was found out that the addition of 1,3-cycloheptanedione to dimethyl acetylenedicarboxylate exhibits a more complex course of the reaction; in the case of 1,3-cyclopentanedione, the product of the *trans* addition does not form 2*H*-pyrone ring, while a derivative of 2*H*-pyrone is the product of *trans* addition of 1,3-cyclohexanediones.

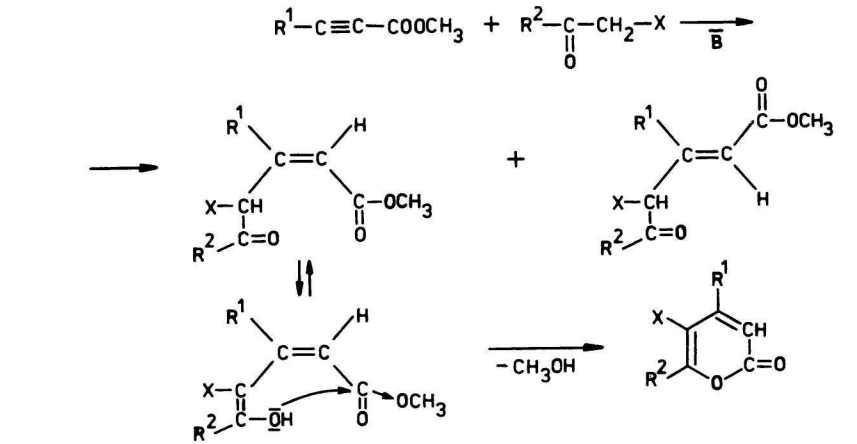
Проведены присоединения по Михаэлю 1,3-циклопентандиона, 1,3-циклогександиона, 5,5-диметил-1,3-циклогександиона и 1,3-циклогептандиона к диметилацетилендикарбоксилату в присутствии катализаторов CH_3ONa, пиперидина или KF/Al_2O_3. Обнаружено, что подобно присоединениям по двойной связи, Михаэлево присоединение по тройной связи успешно осуществляется при катализе KF/Al_2O_3. Изучено влияние катализатора на соотношение *цис*/*транс* продуктов присоединения. Катализ KF/Al_2O_3 в большей степени ведет предпочтительно к *цис*-присоединению, чем катализ метоксидом натрия. Присоединение 1,3-циклогептандиона к диметилацетилендикарбоксилату протекает по более сложному пути. В случае 1,3-циклопентандиона продукт *транс*-присоединения не образует 2*H*-пироновый цикл, в то время как 2*H*-пироновое производное является продуктом *транс*-присоединения 1,3-циклогександионов.

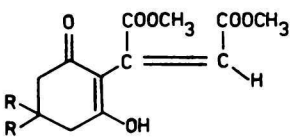
The use of alkali metal fluorides as catalysts in the Michael additions to a C=C bond led to higher yields of products in comparison with the additions catalyzed by alkoxide ions. The yields were particularly increased in those cases when either the higher solubility of fluorides in organic solvents was achieved by the presence of crown ethers, or when fluorides were supported on inorganic carriers, especially on Al_2O_3 [1—4].

Since Michael additions to a triple bond catalyzed by KF/Al_2O_3 have not been accomplished yet, we have performed these reactions and compared the yields achieved with the yields of additions catalyzed by sodium methoxide or piperidine. Dimethyl acetylenedicarboxylate was used as an acceptor in the Michael additions and following 1,3-cycloalkanediones served as donors: 1,3-cyclopentanedione, 1,3-cyclohexanedione, 5,5-dimethyl-1,3-cyclohexanedione, and 1,3-cycloheptanedione. The choice of the above 1,3-cycloalkanediones simultaneously enabled us to examine also the influence of a ring size upon the character of additions products, as in one of our previous works [5] we have found out that the mentioned 1,3-cycloalkanediones as donors and 1-(X-phenyl)-2-nitroethenes as acceptors gave different final products depending on a ring size of a 1,3-cycloalkanedione.

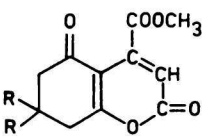
When an acceptor possessing a triple bond reacts in Michael addition with a donor in 1 1 mole ratio, one can assume the formation of *E* and *Z* isomers, in accord with *cis* or *trans* addition of the donor. Current opinions on the knowledge about the stereochemistry of such additions are ambiguous. Some authors, *e.g. Truce* and *Tikhonov* [6], have found the addition of thiophenols to compound of the type $HC{\equiv}C{-}X$ ($X = CN$, SO_2Ar, $C_6H_4NO_2$) to be exclusively *trans*, while *Khetan* and *George* [7] have described the addition of phenacyl amines to dimethyl acetylenedicarboxylate as *cis* addition. Since papers dealing with Michael additions involving triple bonds (and with the stereochemistry of the addition in particular) are rare, one is not able to predict whether selective *cis* or *trans* addition will take place in our case, or both isomers will be formed. If an ester of propiolic acid (or its homologue) is used as an acceptor in the Michael addition and carbonyl compounds are donors, the preliminarily formed *trans* addition intermediate can give rise, after elimination of a molecule of alcohol, to a derivative of 2*H*-pyrone [8—10]. Accordingly, one can also expect formation of 2*H*-pyrone derivatives in the case of the *trans* addition of 1,3-cycloalkanediones to dimethyl acetylenedicarboxylate (Scheme 1).

As 5,5-dimethyl-1,3-cyclohexanedione (dimedone) is commercially available, we used this compound as the first donor among all the above-mentioned 1,3-cycloalkanediones. The addition to dimethyl acetylenedicarboxylate was catalyzed by sodium methoxide, KF/Al_2O_3 or piperidine under conditions given in Experimental. Monitoring of the course of the reaction by TLC revealed formation of several compounds. Column chromatography of the reaction mixture on silica gel (eluant benzene—acetone, $\varphi_r = 8$ 1) separated two chief reaction products, *i.e.* the product of the *cis* addition (*I-Z*) as well as the product of the *trans* addition; the latter, however, was isolated as the derivative of 2*H*-pyrone (*I-E*). Michael addition of dimedone to dimethyl acetylenedicarboxylate was also carried out at 25 °C, with prevailing formation of the *cis* addition product as a result. When the reaction was conducted in refluxing

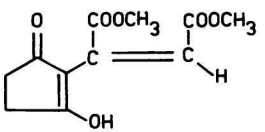




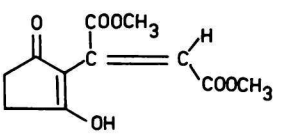
R = CH_3 *I-Z*
R = H *II-Z*



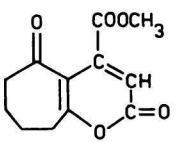
R = CH_3 *I-E*
R = H *II-E*



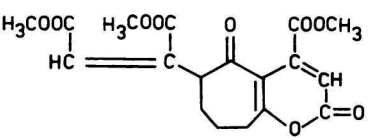
III-Z



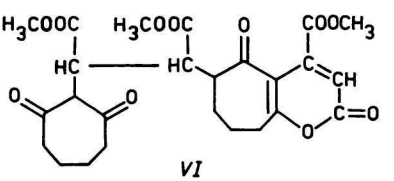
III-E



IV



V



VI

Scheme 1

methanol the *trans* addition product predominated (Table 1). The reaction catalyzed by piperidine gave only 24 % overall yield of mainly *cis* addition product. Since modification of the reaction conditions (time, temperature, solvent) did not improve the yields, piperidine was rejected as a catalyst in Michael additions of other 1,3-cycloalkanediones. The best yields were obtained under KF/Al_2O_3 catalysis. Also in this case the reaction was monitored by TLC which indicated that in refluxing acetonitrile the reaction was over in 30 min.

Table 1

The results of Michael additions of 1,3-cycloalkanediones to dimethyl acetylenedicarboxylate

Donor	Method	Time/h	Yield/%	Ratio of isomers *trans* : *cis*
1,3-Cyclopentanedione	*A*	3	25	1 : 0
	B	0.5	61	1.6 1
1,3-Cyclohexanedione	*A*	5	52	2 1
	B	0.5	68	1 1.4
5,5-Dimethyl-1,3-cyclohexanedione	*A*	5	48	1.4 1
	B	0.5	70	1 1.7

Method *A*: CH_3ONa, refluxing methanol.
Method *B*: KF/Al_2O_3, refluxing acetonitrile.

The overall yield (70 %) was higher than that achieved by sodium methoxide method, but the product of the *cis* addition was a major one (Table 1). The determination of the structure of both *cis* and *trans* addition products was rather easy due to the fact that the *trans* addition product was always isolated in the form of 2*H*-pyrone derivatives, the spectral characteristics (^1H NMR, IR) of which are entirely different from those of the *cis* addition product (see spectral data in Experimental). The addition of 1,3-cyclohexanedione to dimethyl acetylenedicarboxylate gave analogous results. In this case too, sodium methoxide in refluxing methanol gave mainly the *trans* addition product (again isolated as 2*H*-pyrone derivative (*II-E*)) and the product of the *cis* addition (*II-Z*) was isolated in lower yield. On the other hand, the reaction catalyzed by KF/Al_2O_3 preferred formation of the product *II-Z* (Table 1). In case of 1,3-cyclohexanedione, the primary product of the *trans* addition was not isolated either, even at the temperature of 25 °C. Similarly to dimedone, the overall yield of the reaction catalyzed by KF/Al_2O_3 was higher than the yield obtained by sodium methoxide method.

The Michael addition of 1,3-cyclopentanedione to dimethyl acetylenedicarboxylate catalyzed by sodium methoxide led to the isolation of just one product

III-E which is the result of the *trans* mode of addition. The reasons for the attribution of this configuration to the compound *III-E* will be given later. The compound *III-E* precipitated from the reaction mixture as a salt from which *III-E* was obtained by dissolving it in water and acidification with hydrochloric acid to pH = 2. When the addition was carried out in acetonitrile under KF/$/Al_2O_3$ catalysis, both stereoisomers were obtained. The compound *III-E* precipitated from the mixture in the same way as it separated under sodium methoxide catalysis, *i.e.* as a salt insoluble in common organic solvents but soluble, as already mentioned, in water; acidification of its aqueous solution gave *III-E* (m.p. = 166—168 °C). The other isomer *III-Z*, formed by the *cis* addition, was obtained by chromatographic separation of the reaction mixture (silica gel, eluant benzene—acetone, $\varphi_r = 8$ 1).

The determination of the structure of the *cis* and *trans* addition products in the case of dimedone or 1,3-cyclohexanedione was simplified by the fact that the *trans* addition products were converted to the 2*H*-pyrone derivatives *I-E*, *II-E* the spectral characteristics of which completely differ from the spectral characteristics of the compounds *I-Z* and *II-Z*. This is not the case in the Michael addition of 1,3-cyclopentanedione to dimethyl acetylenedicarboxylate. Hence, determining the structure of the *cis* and the *trans* addition products *III-Z*, *III-E* is more complex, as both compounds are mere isomers. Their ^1H and ^{13}C NMR, and IR spectra show only few differences. In ^1H NMR spectra, the compound *III-E* exhibits a singlet at $\delta = 2.41$ ppm (4H) attributed to methylene protons of the cyclopentanone moiety, two singlets at $\delta = 3.55$ and 3.63 ppm (2 × 3H) were assigned to methyl protons of the ester groups and a singlet at $\delta = 6.79$ ppm (1H) to the proton of =CH— group. In ^1H NMR spectra of the isomer *III-Z*, a singlet of methylene protons of the cyclopentanone moiety appears at $\delta = 2.16$ ppm (4H), two singlets belonging to methyl protons of the ester groups are at $\delta = 3.50$ and 3.61 ppm (2 × 3H), and a singlet of the =CH— proton is at $\delta = 6.82$ ppm (1H). In IR spectra, the compound *III-Z* has a peak at $\tilde{\nu} = 987$ cm^{-1} which is absent in IR spectra of *III-E*. The rest of the spectral characteristics of *III-E* and *III-Z* are very much alike (see Experimental). In order to characterize the compounds *III-E* and *III-Z* one has to touch on the ability of *III-E* to form a red complex compound with $FeCl_3$, while *III-Z* is inert. Perhaps melting point of *III-Z* (220—225 °C) too, speaks in favour of the geometry attributed to *III-Z*, as the compounds *I-Z* and *II-Z* have similar melting points. For completeness' sake, we would like to mention that none of the isomers *III-E*, *III-Z* was converted to the derivative of 2*H*-pyrone by heating at 200 °C.

In comparison with preceding 1,3-cycloalkanediones, the Michael addition involving 1,3-cycloheptanedione as a donor proved to be more complicated. The reaction mixture turned black in several minutes irrespective of the catalyst

used — sodium methoxide or KF/Al_2O_3. TLC monitoring of the reaction indicated formation of a mixture of compounds with, largely, close R_f values. From such a multiplicity of compounds we were able to isolate in pure state only products with a higher relative molecular mass using column chromatography on silica gel (benzene—acetone, $\varphi_r = 8$ 1) followed by rechromatography. These compounds are products of successive Michael additions and, on the basis of mass, ^{1}H NMR, and IR spectra, structures of compounds *V* and *VI* were ascribed to them. The compound *V* was formed by the Michael addition of methyl 2,5-dioxo-2,5,6,7,8,9-hexahydrocyclohepta[*b*]pyran-4-carboxylate (*IV*) to dimethyl acetylenedicarboxylate. The compound *IV* arose from the *trans* addition of 1,3-cycloheptanedione to dimethyl acetylenedicarboxylate, followed by cleavage of a methanol molecule. One has to mention that we did not succeed in isolation of *IV* in a pure state. The compound *V* gave rise to *VI* by addition of a second molecule of 1,3-cycloheptanedione. In addition to *V* and *VI*, a compound with the relative molecular mass of 458 was isolated. This compound, however, is not regarded as a product of the primary Michael addition. Probably it was formed from compound *VI*. The determination of the structures of compounds *V* and *VI* resulted from the analysis of their ^{1}H NMR, IR, and mass spectra.

The present experimental results do not allow to draw any conclusions about *cis* addition of 1,3-cycloheptanedione to dimethyl acetylenedicarboxylate. Neither the primary product of the *cis* addition nor products of subsequent reactions which would indicate such a course of the reaction were isolated. Our effort to trap the primary addition products by modification of the reaction conditions (shorter reaction time, change of the catalyst and solvent) has failed.

Experimental

^{1}H NMR spectra were recorded on a Tesla BS 487 instrument with 80 MHz working frequency in C^2HCl_3 or DMSO solutions with TMS as an internal standard. IR spectra were taken on a Specord 75 IR instrument in the region of $\tilde{\nu} = 400$—4000 cm^{-1}. Polystyrene was used as a standard to calibrate its scale. Mass spectra were measured on an MS 902 S (AEI Manchester) instrument at ionizing energy 70 eV. The catalyst KF/Al_2O_3 was prepared by a published procedure [11].

Addition of 5,5-dimethyl-1,3-cyclohexanedione and 1,3-cyclohexanedione to dimethyl acetylenedicarboxylate. Preparation of compounds I-Z, I-E, II-Z, II-E

Method A (CH_3ONa *catalysis*)

To a solution of CH_3ONa, prepared from sodium (0.048 g; 2 mmol) and methanol (5 cm^3), the respective diketone (15 mmol) and dimethyl acetylenedicarboxylate

(15 mmol) is added. The reaction mixture is stirred and refluxed for 5 h and left to stand at 0 °C for 4 days. The crude product is subjected to column chromatography on silica gel (benzene—acetone, $\varphi_r = 8$ 1). Pyrone derivative *I-E*, *II-E* is eluted first, followed by the compounds *I-Z*, *II-Z*. (Further fractions contained several compounds which were not identified due to their close R_f values.) The yields are given in Table 1.

Method B (KF/Al_2O_3 *catalysis*)

To a solution of the respective diketone (11 mmol) in dry acetonitrile (50 cm^3) dimethyl acetylenedicarboxylate (11 mmol) and KF/Al_2O_3 (3.0 g; 20 mmol) [11] is added. The reaction mixture is stirred and refluxed for 30 min, filtered while hot and the solids are washed with acetonitrile (3 × 5 cm^3). The filtrate is concentrated to half of its original volume and allowed to crystallize for 3 days. The crystals separated (*I-Z*, *II-Z*) are filtered off and crystallized from acetone. The filtrate is chromatographed as in the method *A*. The yields are given in Table 1.

Methyl 2,5-dioxo-7,7-dimethyl-5,6,7,8-tetrahydro-2H-chromene-4-carboxylate (I-E): M.p. = 98—99 °C (cyclohexane). For $C_{13}H_{14}O_5$ ($M_r = 250.25$) w_i(calc.): 62.43 % C, 5.61 % H; w_i(found): 62.50 % C, 5.64 % H. ^1H NMR spectrum (C^2HCl_3), δ/ppm: 1.10 (s, 6H, CH_3), 2.41 (s, 2H, CH_2), 3.86 (s, 3H, $COOCH_3$), 6.16 (s, 1H, =CH—). IR spectrum ($CHCl_3$), $\tilde{\nu}/cm^{-1}$: 1747, 1680 (ν(C=O)), 1627 (ν(C=C)).

Dimethyl Z-(4,4-dimethyl-2-hydroxy-6-oxo-1-cyclohexen-1-yl)butenedioate (I-Z): M.p. = 240—245 °C (decomp.) (acetone). For $C_{14}H_{18}O_6$ ($M_r = 282.30$) w_i(calc.): 59.57 % C, 6.38 % H; w_i(found): 59.24 % C, 6.23 % H. ^1H NMR spectrum (DMSO), δ/ppm: 0.82 (s, 6H, CH_3), 1.95 (s, 4H, CH_2), 3.37 (s, 3H, $COOCH_3$), 3.47 (s, 3H, $COOCH_3$), 7.10 (s, 1H, =CH—). IR spectrum (nujol), $\tilde{\nu}/cm^{-1}$: 1707, 1680 (ν(C=O)), 1594 (ν(C=C)).

Methyl 2,5-dioxo-5,6,7,8-tetrahydro-2H-chromene-4-carboxylate (II-E): M.p. = = 101—103 °C (cyclohexane). For $C_{11}H_{10}O_5$ ($M_r = 238.00$) w_i(calc.): 59.46 % C, 4.54 % H; w_i(found): 58.99 % C, 4.42 % H. ^1H NMR spectrum (C^2HCl_3), δ/ppm: 2.12 (m, 2H, CH_2), 2.55 (t, 2H, CH_2), 2.85 (t, 2H, CH_2), 3.87 (s, 3H, $COOCH_3$), 6.13 (s, 1H, =CH—). IR spectrum ($CHCl_3$), $\tilde{\nu}/cm^{-1}$: 1747, 1680 (ν(C=O)), 1625 (ν(C=C)).

Dimethyl Z-(2-hydroxy-6-oxo-1-cyclohexen-1-yl)butenedioate (II-Z): M.p. = 225—230 °C (decomp.) (acetone). For $C_{12}H_{14}O_6$ ($M_r = 254.23$) w_i(calc.): 56.69 % C, 5.50 % H; w_i(found): 56.14 % C, 5.48 % H. ^1H NMR spectrum (DMSO), δ/ppm: 1.62 (m, 2H, CH_2), 2.08 (m, 4H, CH_2), 3.38 (s, 3H, $COOCH_3$), 3.48 (s, 3H, $COOCH_3$), 7.01 (s, 1H, =CH—). IR spectrum (nujol), $\tilde{\nu}/cm^{-1}$: 1707, 1693 (ν(C=O)), 1613 (ν(C=C)).

Addition of 1,3-cyclopentanedione to dimethyl acetylenedicarboxylate Preparation of compounds III-E, III-Z

Method A (CH_3ONa *catalysis*)

To a solution of CH_3ONa, prepared from sodium (0.048 g; 2 mmol) and methanol (5 cm^3), 1,3-cyclopentanedione (1.47 g; 15 mmol) and dimethyl acetylenedicarboxylate

(2.13 g; 15 mmol) is added and the mixture is stirred and refluxed for 3 h. The solution becomes dark and a compound insoluble in common organic solvents but soluble in water separates. It is dissolved in water and the aqueous solution is acidified with hydrochloric acid to pH = 2. The compound *III-E* is filtered off, dried and purified by washing with chloroform. The yield is given in Table 1.

The methanolic filtrate is concentrated to half of its volume and left to stand for 4 days. The oil that separated is composed of several compounds with close R_f values. The structure of these compounds was not identified.

Method B (KF/Al_2O_3 *catalysis)*

In a three-necked flask equipped with a mechanical stirrer and a condenser with a drying tube is placed: 1.07 g (11 mmol) of 1,3-cyclopentanedione, 50 cm^3 of acetonitrile, 1.62 g (11 mmol) of dimethyl acetylenedicarboxylate, and 3 g of KF/Al_2O_3. Contents of the flask is stirred and heated to reflux for 40 min, during which time it turns brown and solid separates. The mixture is hot-filtered and the solid washed with acetonitrile (3×5 cm^3). The filtrate is concentrated to half of its volume and left to stand for 3 days. The compound *III-Z* which separates is filtered off and crystallized from chloroform.

The solid obtained from the first filtration contains KF/Al_2O_3 and compound *III-E*. Treatment with water and acidification of aqueous extracts leads to isolation of *III-E*, identical with the compound isolated in the method *A*. The yields are given in Table 1.

Dimethyl E-(2-hydroxy-5-oxo-1-cyclopenten-1-yl)butenedioate (III-E) M.p. = 166 —168 °C. For $C_{11}H_{12}O_6$ (M_r = 240.21) w_i(calc.): 54.95 % C, 4.99 % H; w_i(found): 54.41 % C, 4.92 % H. ^{1}H NMR spectrum (DMSO), δ/ppm: 2.41 (s, 4H, CH_2), 3.55 (s, 3H, $COOCH_3$), 3.63 (s, 3H, $COOCH_3$), 6.78 (s, 1H, =CH—). IR spectrum (nujol), $\tilde{\nu}$/cm^{-1}: 1733, 1720 (ν(C=O)), 1600 (ν(C=C)), 1427 (ϱ(CH)), 627 (ω(CH)).

Dimethyl Z-(2-hydroxy-5-oxo-1-cyclopenten-1-yl)butenedioate (III-Z) M.p. = 220 —225 °C (decomp.) ($CHCl_3$). For $C_{11}H_{12}O_6$ (M_r = 240.21) w_i(calc.): 54.95 % C, 4.99 % H; w_i(found): 54.32 % C, 4.85 % H. ^{1}H NMR spectrum (DMSO), δ/ppm: 2.16 (s, 4H, CH_2), 3.50 (s, 3H, $COOCH_3$), 3.61 (s, 3H, $COOCH_3$), 6.82 (s, 1H, =CH—). IR spectrum (nujol), $\tilde{\nu}$/cm^{-1}: 1720, 1693 (ν(C=O)), 1593 (ν(C=C)), 987 (τ(CH)).

Addition of 1,3-cycloheptanedione to dimethyl acetylenedicarboxylate Preparation of compounds V VI

The addition is performed in the same way as the additions of 1,3-cyclohexanedione and dimedone (methods *A* and *B*). As during 4 days no separation of solid is observed in none of the methods, the reaction mixture is subjected to column chromatography on silica gel (benzene—acetone, φ_r = 8 1). The eluant is concentrated and the residue rechromatographed on silica gel (benzene—dichloromethane, φ_r = 2 1). The compound *V* is eluted first, followed by the compound *VI*.

Dimethyl (4-methoxycarbonyl-2,5-dioxo-2,5,6,7,8,9-hexahydrocyclohepta[b]pyran-6-yl)butenedioate (V) Yield = 14 %. M.p. = 131—132 °C (cyclohexane). $M^+ = 380$. For $C_{18}H_{20}O_9$ ($M_r = 380.23$) w_i(calc.): 56.84 % C, 5.26 % H; w_i(found): 56.32 % C, 4.99 % H. ^{1}H NMR spectrum (C^2HCl_3), δ/ppm: 2.10 (m, 2H, CH_2), 2.70 (t, 2H, CH_2), 2.90 (t, 2H, CH_2), 3.89 (s, 3H, $COOCH_3$), 3.96 (s, 3H, $COOCH_3$), 4.02 (s, 3H, $COOCH_3$), 6.27 (t, 1H, =CH—), 7.28 (s, 1H, =CH—). IR spectrum ($CHCl_3$), $\tilde{\nu}$/cm^{-1}: 1787, 1747 (ν(C=O)), 1600 (ν(C=C)).

Dimethyl 1-(1,3-dioxo-2-cycloheptyl)-2-(4-methoxycarbonyl-2,5-dioxo-2,5,6,7,8,9-hexahydrocyclohepta[b]pyran-6-yl)butanedioate (VI): Yield = 7 %. M.p. = 230—236 °C. $M^+ = 504$. For $C_{25}H_{28}O_{11}$ ($M_r = 504.30$) w_i(calc.): 59.52 % C, 5.55 % H; w_i(found): 59.61 % C, 5.61 % H. ^{1}H NMR spectrum (C^2HCl_3), δ/ppm: 1.90 (m, 8H, CH_2), 2.60 (m, 4H, CH_2), 2.90 (m, 2H, CH_2), 3.62 (s, 3H, $COOCH_3$), 3.74 (s, 3H, $COOCH_3$), 3.88 (s, 3H, $COOCH_3$), 4.30 (m, 4H, =CH—), 7.30 (s, 1H, =CH—). IR spectrum (nujol), $\tilde{\nu}$/cm^{-1}: 1787, 1765, 1748 (ν(C=O)), 1605 (ν(C=C)).

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