

Preparation and some properties of 1,3-cyclotridecanedione and 1,3-cyclotetradecanedione

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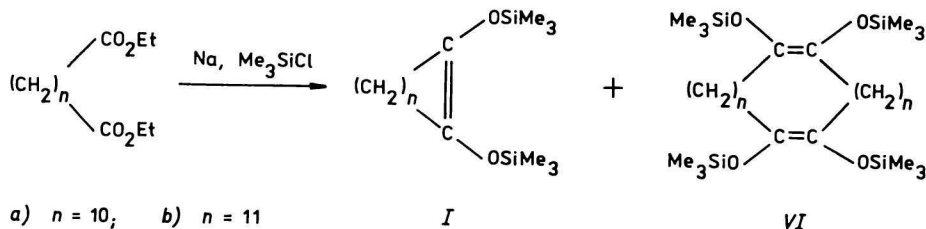
1,3-Cyclotridecanedione (*IIIa*) and 1,3-cyclotetradecanedione (*IIIb*) have been prepared by a three-step synthesis from diethyl dodecanedioate and tridecanedioate respectively, and the structure of by-products of first two reactions has been proved. By means of $^1\text{H-n.m.r.}$ and infrared spectroscopy it has been found out that *IIIa* and *IIIb* exist in the ketoenol form which is stabilized by intramolecular hydrogen bond. Methylation of the anions of *IIIa* and *IIIb* took place at the carbon atoms in low yields. Reaction of *IIIa* with diazomethane did not afford the expected product of *O*-methylation but 2,2'-methylenedi-1,3-cyclotridecanedione (*VII*) was isolated.

1,3-Циклотридекандион (*IIIa*) и 1,3-циклотетрадекандион (*IIIb*) были синтезированы путем трехстадийного синтеза из диэтилдодекандиоата и -тридекандиоата соответственно, причем была установлена структура побочных продуктов первых двух стадий синтеза. Методами $^1\text{H-NMR}$ и ИК-спектроскопии было показано, что *IIIa* и *IIIb* существуют в кетоенольной форме, стабилизированной внутримолекулярной водородной связью. Метилирование анионов *IIIa* и *IIIb* протекает по атому углерода с низкими выходами. Реакция *IIIa* с диазометаном не привела к предполагаемому продукту *O*-метилирования, но был выделен 2,2'-метиленди-1,3-циклотридекандион (*VII*).

There are not many reports on preparation of 1,3-cycloalkanediones with eight and more membered rings. In 1966 *Schank* and *Eisert* [1] described a general method for the preparation of 1,3-cycloalkanediones with more than seven carbon atoms in the ring. The utility of this method is diminished however, by the lack of availability of starting cyclic ketones and low total yields (3—20 %). Another approach to 1,3-cycloalkanediones substituted in position 2 was published by *Hünig et al.* [2] who exploited cycloalkanones and acid chlorides as starting material. The possibility of preparation of 1,3-cyclotridecanedione by this method is also mentioned there.

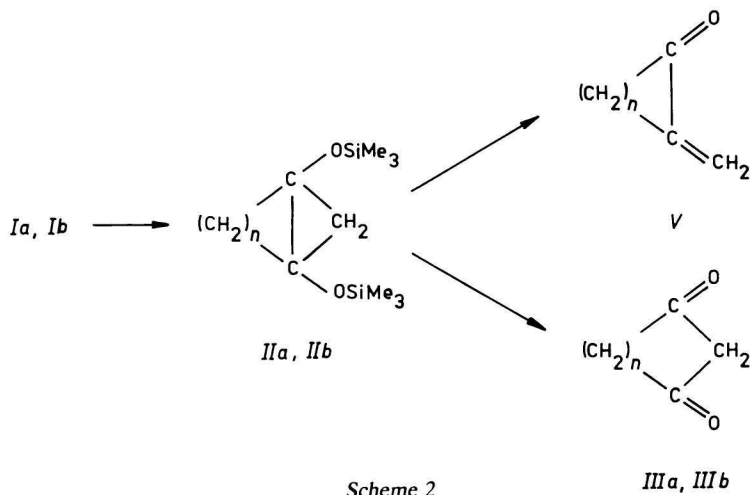
We have already described a procedure for the preparation of 1,3-cycloalkanediones with seven through eleven carbon atoms in the ring [3]. This method,

originally developed for the preparation of 2-cycloalkene-1-ones [4], was later applied in the synthesis of 1,3-cyclopentadecanedione by the same author [5]. We have adapted this procedure also for the preparation of 1,3-cyclotridecanedione (*IIIa*) and 1,3-cyclotetradecanedione (*IIIb*). The first step of the synthesis is the well known acyloin condensation of dicarboxylic acid diesters in the presence of trimethylchlorosilane [6, 7]. This reaction proceeds in high yields (80–90 %) when small and common rings (4–7 carbon atoms) are being closed. We obtained yields of 40–60 % in the case of *Ia* and *Ib* (Scheme 1) under high dilution conditions, using xylene as the solvent and with excluding moisture and oxygen. In toluene, under the same conditions, *Ia* and *Ib* were formed in 25–30 % yields only, whereas *Bloomfield et al.* [7] reported as high as 60–70 % yields. This decreasing can be explained by a competitive intermolecular acyloin condensation which is followed by the 24- or 26-membered ring closure. Such compounds (*VI*) were isolated from the reaction mixture.



Scheme 1

The second step, Simmons—Smith reaction, was performed on *Ia* and *Ib* in two ways. At first, diiodomethane and zinc activated by copper(I) chloride were used as the reagent. The advantage of this couple consists in high and reproducible yields and therefore it has been widely used in cyclopropanation reactions since its discovering. In spite of this, 1,12-bis(trimethylsiloxy)bicyclo[10.1.0]tridecane (*IIa*) and 1,13-bis(trimethylsiloxy)bicyclo[11.1.0]tetradecane (*IIb*) were formed in poor yields under these conditions. It is known [8] that substitution of zinc-copper couple by diethylzinc often enables to perform successfully such cyclopropanation reactions that failed with the former reagent. Taking into account the properties of diethylzinc (pyrophoric) we preferred an ethereal solution of ethylzinciodide, the reagent in Simmons—Smith reaction suggested by *Sawada and Inouye* [9]. Treatment of this reagent and diiodomethane with *Ia* and *Ib* led to cyclopropane derivatives *IIa* and *IIb* (Scheme 2) in 74 % yields. Carrying out these reactions one should be careful however, as zinc iodide, the by-product of the reaction, is able to cleave the three-membered ring of compounds *II* [10]. Prevention from this undesirable complication is relatively high dilution of the reactants and scrupulous removing of the zinc salts after reaction is complete. 2-Methylenecyclododecanone



(V) was the main product when cyclopropanation of *Ia* was conducted at a concentration higher than that given in Experimental.

Compounds *IIa* and *IIb* were identified by their i.r. and ¹H-n.m.r. spectra. The i.r. spectra of *IIa* and *IIb* show no peak of C=C bond stretching vibration in comparison with i.r. spectra of starting compounds *Ia* and *Ib*. On the other hand, their ¹H-n.m.r. spectra exhibit the signals of the protons of the methylene group which is a part of the three-membered ring. The compound V has a weak peak in the region of stretching vibration of the carbonyl group at $\tilde{\nu} = 1780 \text{ cm}^{-1}$ and a strong one in the region of stretching vibration of C=C bond at $\tilde{\nu} = 1675 \text{ cm}^{-1}$, which is typical for the mutual *cis* arrangement of these groups. The chemical shifts of the protons of methylene group (=CH₂) are at $\delta_r = 5.53 \text{ ppm}$ and 5.67 ppm .

The third step of the synthesis consists in the oxidation of *IIa* and *IIb* by anhydrous iron(III) chloride in DMF. Desired diketones *IIIa* and *IIIb* were prepared in 65 % yields. Their structure was determined by both spectral methods and elemental analysis. The ¹H-n.m.r. spectra show not only the signals of the protons of methylene group between two carbonyls ($\delta_r = 3.60 \text{ ppm}$) but the signal of the proton which belongs to the =CH—CO— grouping ($\delta_r = 5.70 \text{ ppm}$) as well. This is an evidence that these compounds exist in both diketo and ketoenol forms. The same piece of information results also from their i.r. spectra. There are three peaks in the region of $\tilde{\nu} = 1500\text{—}1800 \text{ cm}^{-1}$; the peak at $\tilde{\nu} = 1610 \text{ cm}^{-1}$ (*IIIa*) or 1595 cm^{-1} (*IIIb*) corresponds to the >C=CH—CO— grouping, while the peaks at $\tilde{\nu} = 1705 \text{ cm}^{-1}$ or 1715 cm^{-1} ($\tilde{\nu}_{\text{as}}(\text{C=O})$) and at $\tilde{\nu} = 1767 \text{ cm}^{-1}$ or 1780 cm^{-1} ($\tilde{\nu}_{\text{s}}(\text{C=O})$) belong to the diketo form. It is worth mentioning the extremely high difference between the wavenumbers of $\tilde{\nu}_{\text{s}}(\text{C=O})$ and $\tilde{\nu}_{\text{as}}(\text{C=O})$ ($\tilde{\nu} = 62$ or 65 cm^{-1}) as well as the fact that the peak of $\tilde{\nu}_{\text{as}}(\text{C=O})$ is stronger than that of

$\bar{\nu}_s(\text{C}=\text{O})$ (the reverse is truth in the case of 1,3-cycloalkanediones with the smaller rings and moreover $\Delta\bar{\nu} = \bar{\nu}_s(\text{C}=\text{O}) - \bar{\nu}_{as}(\text{C}=\text{O})$ reaches only 19–33 cm^{-1} [3]). These facts indicate that diketo form of *IIIa* and *IIIb* possesses *U* conformation (Fig. 1), in contrast with lower 1,3-cycloalkanediones which can exist, owing to their rigidity, exclusively in *W* conformation. The size and flexibility of 13- and 14-membered rings enables to form any of the mentioned conformational arrangements, as it was checked by both Dreiding and space-fillings models. The ketoenol form of compounds *IIIa* and *IIIb* is stabilized by internal hydrogen bonding which is unavailable to *W* conformation. Therefore, the structure of 1,3-cyclotridecanedione and 1,3-cyclotetradecanedione resembles rather the structure of aliphatic 1,3-diketones than that of their lower homologues.

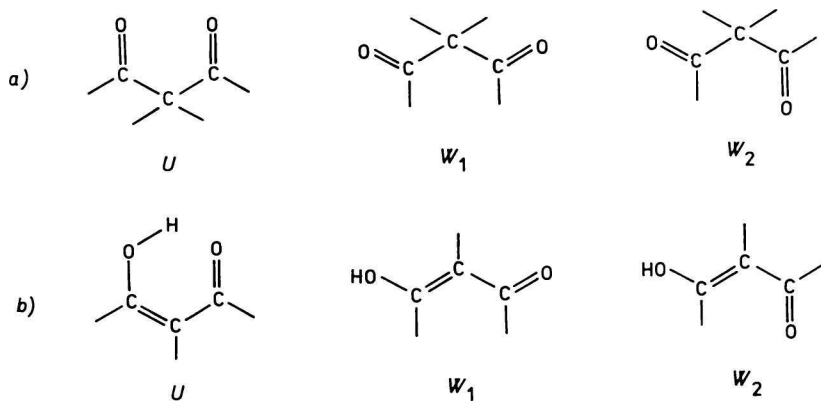


Fig. 1. Conformational arrangements of diketo form (a) and ketoenol form (b) of β -dicarbonyl compounds.

The ketoenol form content of *IIIa* and *IIIb* was determined in various solvents by means of ^1H -n.m.r. spectroscopy. The spectra were taken after the equilibrium in a given solvent had been reached (one week standing at 24 $^\circ\text{C}$). For comparison, the same dependence was studied also for 2,4-pentanedione. As seen from Table 1, the ketoenol content of all studied compounds decreases with the increased polarity of the solvent. This result confirms our assumption of the identical conformational arrangements of *IIIa*, *IIIb*, and acyclic β -diketones. The highest ketoenol form of all three studied diketones was found in pyridine, which is surely caused by the basicity of this solvent. It is also evident that *IIIa* has a smaller ketoenol content than *IIIb* in any of solvents.

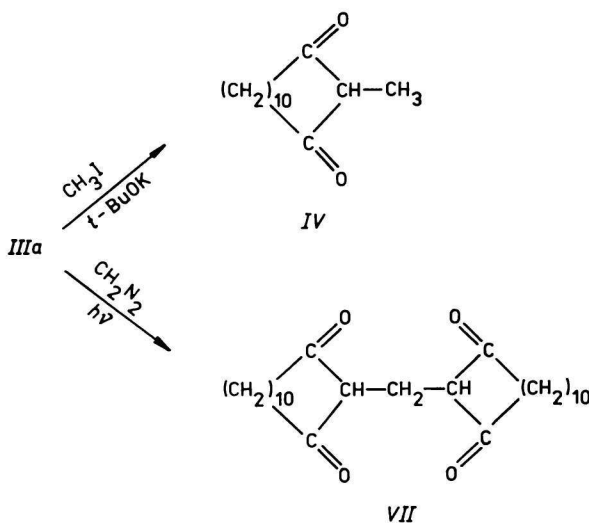
Our previous work [11–15] resulted in finding out that the polarity of the solvent used in the alkylation of 1,3-cycloalkanedione anions affects the ratio of *C*- and *O*-alkylated products in just opposite way as it is in the case of the anions of

Table 1

The ketoenol form content of 2,4-pentanedione (*VIII*), 1,3-cyclotridecanedione (*IIIa*), and 1,3-cyclo-tetradecanedione (*IIIb*) in various solvents at 24 °C

Deuterated solvent	Ketoenol form content/%		
	<i>VIII</i>	<i>IIIa</i>	<i>IIIb</i>
Chloroform	80	76	88
Pyridine	92	80	92
Acetone	75	67	71
Acetonitrile	63	60	63
Dimethyl sulfoxide	55	45	50

aliphatic β -dikarbonyl compounds. Trying to find out the influence of the solvent nature on the C/O ratio, we run the alkylation of the anions of *IIIa* and *IIIb* with iodomethane in different solvents. It was observed however, that product of *O*-methylation was not formed in any of them (DMF, DMSO, acetone, 2-methyl-2-propanol) and only *C*-methylated derivative *IV* (Scheme 3) was isolated in low yields. The attempts to prepare *O*-methylated derivatives with diazomethane have also failed. There was no reaction at temperatures of 0–5 °C (in a refrigerator) and starting compounds were isolated. When the reaction mixture was left to stand at room temperature, in addition to unchanged *IIIa*



Scheme 3

a compound with high melting point (154—155 °C) was isolated. The structure of this compound determined by the mass, ^1H -n.m.r., i.r. spectra, and elemental analysis is that of VII (Scheme 3). Such an anomalous behaviour of the compounds IIIa and IIIb can be attributed either to the formation of strong hydrogen bonds or to a conformational arrangement which involves the $-\text{CO}-\text{CH}_2-\text{CO}-$ or $-\text{C}(\text{OH})=\text{CH}-\text{CO}-$ grouping turned in the ring.

Experimental

Melting points were determined on a Kofler hot stage. Infrared spectra were recorded on a Perkin—Elmer 567 spectrophotometer in the region of $\tilde{\nu}=400-4000\text{ cm}^{-1}$ in CCl_4 solutions and using NaCl cells. The ^1H -n.m.r. spectra were taken on a Tesla BS 487 instrument operating at 80 MHz, with tetramethylsilane as internal standard and CDCl_3 as solvent. Dodecanedioic acid was prepared by the modification of the procedure for preparation of docosanedioic acid [16] (ester chloride of adipic acid was used as acylating agent instead of adipic dichloride). Tridecanedioic acid was prepared from 2,2'-methylene-1,3-cyclohexanedione [17]. Their diethyl esters were prepared according to Mićović [18].

1,2-Bis(trimethylsiloxy)cycloalkenes (*Ia*, *Ib*)

Trimethylchlorosilane (1.20 mol) is mixed with the cooled (50 °C) suspension of sodium (1.10 mol) in dry xylene (900 cm^3) under dry, oxygen-free nitrogen. A solution of diethyl alkanedioate (0.25 mol) in xylene (400 cm^3) is added during 24 h to the stirred suspension at the temperature of 80—85 °C (an oil bath). The stirring is then continued for additional 20 h at 100 °C, the reaction mixture is cooled to room temperature, the precipitated sodium chloride is sucked and washed with xylene (5 \times 60 cm^3). The solvent is removed (VRE) from combined filtrates and the residue is rectified on a Vigreux column (20 cm) under reduced pressure, yielding 40—50 % of 1,2-bis(trimethylsiloxy)cyclodecene (*Ia*), b.p. (13 Pa) = 115—120 °C, $n_D^{20} = 1.4688$. The yield of 1,2-bis(trimethylsiloxy)cyclotridecene (*Ib*), b.p. (13 Pa) = 125—130 °C, $n_D^{20} = 1.4701$, is 55—60 %. Both constants are in accord with the literature data [7].

1,2,13,14-Tetrakis(trimethylsiloxy)-1,13-cyclotetracosadiene (*VIa*)

The solid residue, left in distillation of *Ia*, is recrystallized from acetone to give *VIa*, m.p. = 75—76 °C, in 25 % yield.

For $\text{C}_{36}\text{H}_{76}\text{O}_4\text{Si}_4$ ($M_r = 685.4$) w_i (calculated): 63.09 % C, 11.18 % H; w_i (found): 62.75 % C, 11.31 % H. IR: $\tilde{\nu}_s(\text{Si}-\text{O}-\text{C}) = 825\text{ cm}^{-1}$, $\tilde{\nu}_{as}(\text{Si}-\text{O}-\text{C}) = 1011\text{ cm}^{-1}$, $\delta(\text{CH}_2) = 1450\text{ cm}^{-1}$, $\tilde{\nu}(\text{C}=\text{C}) = 1660\text{ cm}^{-1}$, $\tilde{\nu}(\text{CH}_2) = 2850\text{ cm}^{-1}$, $\tilde{\nu}(\text{CH}_3) = 2910\text{ cm}^{-1}$. ^1H -NMR (δ ,/ppm): 1.20 (40 H, s, $-\text{CH}_2-$).

1,n + 2-Bis(trimethylsiloxy)bicyclo[n.1.0]alkanes (IIa, IIb)

Diiodomethane (56 mmol) is added to the solution of ethylzinciodide (0.12 mol) in dry ether (100 cm³) [9] and resulting mixture is held under reflux and stirred in nitrogen atmosphere for 1 h. A solution of 1,2-bis(trimethylsiloxy)cycloalkene (40 mmol) in dry ether (50 cm³) is added over a period of 30 min and the stirring under reflux is kept on for 48 h. The reaction mixture is diluted with dry ether (150 cm³) and the excess of the reagent is quenched with dry pyridine (56 mmol) added during 1 h at the temperature of 0 °C. The content of the flask is warmed to 20 °C, the precipitate is removed by suction filtration and washed with dry ether (3 × 25 cm³). Dry pyridine is added dropwise to the combined filtrates until no more precipitate is formed. After filtration the solvent is removed and the residue is distilled under reduced pressure.

1,12-Bis(trimethylsiloxy)bicyclo[10.1.0]tridecane: Yield 75 %, b.p. (13 Pa) = 128—132 °C, $n_D^{20} = 1.4717$.

For C₁₉H₃₀O₂Si₂ ($M_r = 356.4$) w_i (calculated): 64.04 % C, 11.22 % H; w_i (found): 64.21 % C, 11.20 % H. IR: $\tilde{\nu}(\text{Si—O}) = 762 \text{ cm}^{-1}$, $\tilde{\nu}_s(\text{Si—O—C}) = 845 \text{ cm}^{-1}$, $\tilde{\nu}_{\text{as}}(\text{Si—O—C}) = 1054 \text{ cm}^{-1}$, $\delta(\text{CH}_2) = 1455 \text{ cm}^{-1}$, $\tilde{\nu}(\text{CH}_2) = 2936 \text{ cm}^{-1}$. ¹H-NMR

(δ_i /ppm): 0.45 (1 H, d, $J = 7 \text{ Hz}$, $\text{CH}_2 \begin{array}{c} \diagup \text{C} \\ \diagdown \text{C} \end{array}$), 0.83 (1 H, d, $J = 7 \text{ Hz}$, $\text{CH}_2 \begin{array}{c} \diagup \text{C} \\ | \\ \diagdown \text{C} \end{array}$), 1.30 (20 H, s, —CH₂—).

1,13-Bis(trimethylsiloxy)bicyclo[11.1.0]tetradecane: Yield 81 %, b.p. (13 Pa) = 133—137 °C, $n_D^{20} = 1.4748$.

For C₂₀H₄₂O₂Si₂ ($M_r = 370.7$) w_i (calculated): 64.80 % C, 11.42 % H; w_i (found): 64.69 % C, 11.52 % H. IR: $\tilde{\nu}(\text{Si—C}) = 762 \text{ cm}^{-1}$, $\tilde{\nu}_s(\text{Si—O—C}) = 848 \text{ cm}^{-1}$, $\tilde{\nu}_{\text{as}}(\text{Si—O—C}) = 1055 \text{ cm}^{-1}$, $\delta(\text{CH}_2) = 1456 \text{ cm}^{-1}$, $\tilde{\nu}(\text{CH}_2) = 2938 \text{ cm}^{-1}$. ¹H-NMR

(δ_i /ppm): 0.56 (1 H, d, $J = 7 \text{ Hz}$, $\text{CH}_2 \begin{array}{c} \diagup \text{C} \\ | \\ \diagdown \text{C} \end{array}$), 0.70 (1 H, d, $J = 7 \text{ Hz}$, $\text{CH}_2 \begin{array}{c} \diagup \text{C} \\ | \\ \diagdown \text{C} \end{array}$), 1.26 (22 H, s, —CH₂—).

2-Methylenecyclododecanone (V)

The procedure is identical with the preparation of *IIa*. The reaction was conducted at twofold amount of the reactants.

Yield: 56 %, b.p. (26 Pa) = 117—120 °C, $n_D^{20} = 1.4925$.

For C₁₃H₂₂O ($M_r = 174.2$) w_i (calculated): 78.15 % C, 12.73 % H; w_i (found): 77.91 % C, 12.12 % H. IR: $\delta(\text{CH}_2) = 1440 \text{ cm}^{-1}$, 1465 cm^{-1} , $\tilde{\nu}(\text{C=C}) = 1675 \text{ cm}^{-1}$, $\tilde{\nu}(\text{C=O}) = 1780 \text{ cm}^{-1}$. ¹H-NMR (δ_i /ppm): 1.25 (18 H, s, —CH₂—), 2.55 (2 H, t, —CH₂—CO—), 5.53 (1 H, d, $J = 14 \text{ Hz}$, CH₂=), 5.67 (1 H, d, $J = 14 \text{ Hz}$, CH₂=).

1,3-Cycloalkanediones (IIIa, IIIb)

1,n + 2-Bis(trimethylsiloxy)bicyclo[n.1.0]alkane (55 mmol) is added under nitrogen during 1 h at room temperature to the stirred solution of anhydrous iron(III) chloride (0.12 mol) in dry DMF (170 cm³). The stirring is continued for 3 h at 40 °C and the solution

is left to stand at room temperature overnight. The reaction mixture is poured into diluted hydrochloric acid (3.6 %, 170 cm³), stirred at room temperature for 3 h and the product is taken in chloroform (6 × 55 cm³). Chloroform extracts are washed with hydrochloric acid (3.6 %, 4 × 30 cm³), with water (4 × 40 cm³) and dried with magnesium sulfate. The solvent is removed and the residue is distilled under reduced pressure.

1,3-Cyclotridecanedione: Yield 65 %, b.p. (133 Pa) = 102–104 °C, n_D^{20} = 1.5042.

For C₁₃H₂₂O₂ (M_r = 210.1) w_i (calculated): 74.31 % C, 10.74 % H; w_i (found): 74.31 % C, 10.47 % H. IR: $\tilde{\nu}$ (>C=CH—CO—) = 1610 cm⁻¹, $\tilde{\nu}_{as}$ (C=O) = 1705 cm⁻¹, $\tilde{\nu}_s$ (C=O) = 1767 cm⁻¹. ¹H-NMR (δ ,/ppm): 1.25 (16 H, s, —CH₂—), 2.33 (4 H, m, —CH₂—CO—), 3.60 (2 H, s, —CO—CH₂—CO—), 5.68 (1 H, s, —CH=).

1,3-Cyclotetradecanedione: Yield 64 %, b.p. (266 Pa) = 126–130 °C, m.p. = 30–33 °C.

For C₁₄H₂₄O₂ (M_r = 224.3) w_i (calculated): 74.95 % C, 10.78 % H; w_i (found): 75.00 % C, 10.75 % H. IR: $\tilde{\nu}$ (>C=CH—CO—) = 1595 cm⁻¹, $\tilde{\nu}_{as}$ (C=O) = 1715 cm⁻¹, $\tilde{\nu}_s$ (C=O) = 1780 cm⁻¹. ¹H-NMR (δ ,/ppm): 1.25 (18 H, s, —CH₂—), 2.34 (4 H, m, —CH₂—CO—), 3.44 (2 H, s, —CO—CH₂—CO—), 5.53 (1 H, s, —CH=).

2-Methyl-1,3-cyclotridecanedione (IV)

Potassium (10 mmol) is allowed to react with 2-methyl-2-propanol (6 cm³) and 1,3-cyclotridecanedione (10 mmol) is added to the suspension after cooling to room temperature. Iodomethane (20 mmol) is introduced and content of the flask is heated until neutral reaction mixture. The solvent is removed, the residue is treated with water (20 cm³) and extracted with ether (3 × 20 cm³). Combined extracts are washed with water (3 × 10 cm³), dried and ether is removed. The residue is crystallized from aqueous acetone affording 26 % IV, m.p. = 57–59 °C.

For C₁₄H₂₄O₂ (M_r = 224.3) w_i (calculated): 74.95 % C, 10.78 % H; w_i (found): 74.12 % C, 11.28 % H. IR: $\tilde{\nu}_{as}$ (C=O) = 1680 cm⁻¹, $\tilde{\nu}_s$ (C=O) = 1720 cm⁻¹. ¹H-NMR (δ ,/ppm): 1.10 (19 H, s, —CH₂—, CH₃—), 2.40 (4 H, m, —CH₂—CO—), 3.60 (1 H, k, CH₃—CH<).

2,2'-Methylenedi-1,3-cyclotridecanedione (VII)

The solution of 1,3-cyclotridecanedione (10 mmol) and diazomethane (15 mmol) in dry ether (30 cm³) is left to stand at room temperature not protected from light until its colour disappears. The ether is removed and the residue is crystallized from acetone. Yield: 25 %, m.p. = 154–155 °C.

For C₂₇H₄₄O₄ (M_r = 432.6) w_i (calculated): 74.96 % C, 10.25 % H; w_i (found): 75.37 % C, 10.69 % H. M^+ (m/z = 432). IR: $\tilde{\nu}_{as}$ (C=O) = 1682 cm⁻¹, $\tilde{\nu}_s$ (C=O) = 1720 cm⁻¹. ¹H-NMR (δ ,/ppm): 1.10 (32 H, s, —CH₂—), 2.40 (10 H, m, —CH₂—CO—, >CH—CH₂—CH<), 3.60 (2 H, t, >CH—).

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