SYNTHESIS OF THE N,S-ACETALS OF DIACYL- AND ALKOXYCARBONYL(ACYL)KETENES WITH β-DICARBONYL COMPOUNDS, ORGANIC THIOCYANATES, AND NICKEL(2+) COMPLEXES

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An effective method is proposed for the production of the N,S-acetals of diacylketenes and acyl(alkoxycarbonyl)ketenes, based on the addition of methylene-active β-diketones and β-ketoesters to alkyl and aryl thiocyanates catalyzed by Ni(2+) acetylacetonate. Methods are indicated for the synthetic utilization of diacetylketene N,S-acetals and include their conversion to ketene aminals, chelation with boron compounds, and deacylation of derivatives of monoacetylketene with methanol in the presence of Co(2+) acetate.

Ketene N,S-acetals have been widely used in heterocyclic synthesis [1,2]. At the same time, among reagents of this type the derivatives of diacyl- and alkoxycarbonyl-(acyl)-ketenes have been studied little. For the synthesis of these compounds it is necessary to obtain the ketene S,S-acetals from β-diketones or β-keto esters, carbon bisulfide, and the respective alkylating agents selectively substitute one alkylthio group by an amino group [1]. Disadvantages of this method include its multistage character, the need to use strong bases, the high toxicity of carbon bisulfide, and also restrictions due to the nature of the employed amine.

Another approach is based on the reactions of methylene-active compounds with isothiocyanates, but this method has been used mainly for the synthesis of the N,S-acetals of monoacylketenes [1]. It should also be noted that the above-mentioned methods are probably not suitable for the production of ketene N,S-acetals unsubstituted at the nitrogen atom.

A promising method for the synthesis of compounds of this type is the addition of β-diketones or β-keto esters to readily obtainable organic thiocyanates. The basic possibility of such transformations of methylene-active compounds in the presence of sodium ethoxide was noted in [3] (with reference to a dissertation), but the initial reagents and experimental details were not indicated specifically.

However, this is contradicted by the well known tendency of organic thiocyanates to undergo cleavage at the C–S bond under the influence of nucleophilic reagents [4]. Thus, the reaction of the sodium salt of acetoacetic ester with ethyl thiocyanate was used for the production of ethyl α-cyanoacetoacetate [5]. Our attempts at the addition of acetylacetone or acetoacetic ester to methyl thiocyanate in the absence of catalysts were unsuccessful. The use of increased pressure (up to 10 kbar) also did not give the desired result. In the presence of sodium ethoxide the reaction is accompanied by the release of methyl hydrosulfide and leads to a mixture of products, in which according to PMR the content of diacetylketene N,S-acetal is not greater than 10-20%.

It is known that some metal β-diketonates are capable of catalyzing the addition of methylene-active β-dicarbonyl compounds (DCC) at reactive multiple bonds [6-9]. Recently we showed that $Ni(acac)_2$ effectively catalyzes the reaction of β-dicarbonyl compounds with cyanamides [10-12], which (like thiocyanates) have a cyano group attached to a heteroatom.

It was found that the β-diketones and β-keto esters (I-III) add smoothly to the organic thiocyanates (IV-VIII) under mild conditions in the presence of 5-10 mole % of $Ni(acac)_2$ with the formation of the corresponding N,S-acetals of diacylketenes (N,S-ADK) and alkoxycarbonyl(acyl)ketenes(N,S-AAK) (IX-XV) (see also the preliminary communication [13]).

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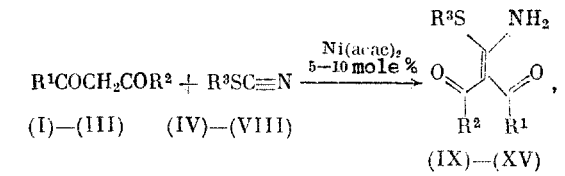


(I)—(III) (IV)—(VIII) (IX)—(XV)

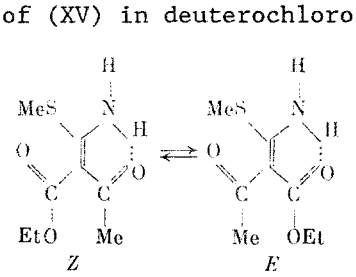
$R^1 = R^2 = Me$ (I), (IX)—(XIII); $R^1 = Me$, $R^2 = Ph$ (II), (XIV), OEt (III), (XV); $R^3 =$ = Me (IV), (IX), (XIV), (XV); Et (V), (X); Pr (VI), (XI); Ph (VII), (XII); *p*-Tol (VIII), (XIII). (1)

Reaction (1) takes place at 20°C in the β-dicarbonyl compound or in THF solution and gives the N,S-ADK and N,S-AAK with yields of 40-87%. Instead of $Ni(acac)_2$ it is possible to use other nickel(2+) β-ketoenolates in this reaction. Although the latter are not commercial products, their use facilitates the production of pure N,S-ADK and N,S-AAK not containing traces of compound (IX), i.e., the product formed from $Ni(acac)_2$. Thus, the synthesis of N,S-ADK (XIV) was realized in the presence of 5 mole% of $Ni(bac)_2$ (bacH is benzoylacetone).

The structure of the synthesized compounds was confirmed by the data from IR and ^1H and ^{13}C NMR spectroscopy and mass spectrometry.

The mass spectra of (IX-XV) contain strong peaks for the molecular ions. The IR spectra contain narrow signals for the free NH (in the region of 3500-3440 cm^{-1}) and the NH involved in intramolecular hydrogen bonds (broad diffuse absorption bands in the region of 3300-2830 cm^{-1}). In the ^{13}C NMR spectra of the N,S-ADK (IX-XIII) in deuterochloroform at 20°C the acetyl groups are equivalent. However, as shown for the case of compound (XIII), with gradual reduction in temperature (-10 to -30°C) their signals become split: 31.70; 32.22 ppm (Me) and 196.07; 198.48 (CO), i.e., the rate of rotation about the C=C bond is reduced.

For the ketene N,S-acetals (XIV) and (XV), which each have two different carbonyl-containing substituents, E/Z isomerism is possible. In the ^1H and ^{13}C spectra of these compounds in deuterochloroform at ~20°C, however, there is only one set of signals. One of the protons of the NH_2 group gives a signal in the region of ~6 ppm, while the other gives a signal in the much more downfield region of ~12 ppm, which indicates that the latter participates in the formation of an intramolecular hydrogen bond. When the solution of (XV) is cooled to -45°C, however, two sets of signals for NH_2 (12.68 and 6.39 ppm, 10.81 and 6.20 ppm) in a ratio of 4:1 are observed. Thus, as in the case of our previously investigated keteneaminals [12] a rapid mutual transformation of the E and the Z isomers on the NMR time scale takes place in the solution of (XV) in deuterochloroform.



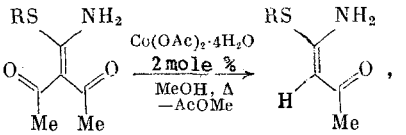
Z *E*

The Z isomer probably predominates, since the possibility of the formation of an intramolecular hydrogen bond with the participation of the acetyl group seems slightly preferred in comparison with the ethoxycarbonyl group (cf. the basicity of acetone and ethyl acetate [14,15]).

The analogous E/Z isomerization process clearly also arises in the case of compound (XIV). The N,S-ADK and N,S-AAK fully substituted at the nitrogen are in general characterized by a very low barrier to rotation about the C=C bond ($\Delta G^{\neq} < 6$ kcal/mole [16]). This is explained by the substantial reduction of its double character in the push-pull system. In the case of compounds (XIV, XV) this barrier should be higher on account of the formation of the intramolecular hydrogen bond (cf. [12,17]).

Earlier we showed that the deacylation of the aminals of diacylketenes by methanol, catalyzed by $Co(OAc)_2$, can be used for the production of the respective derivatives of monoacylketenes [19].

It was found that the N,S-ADKs undergo a similar transformation when boiled in methanol with 2 mole % of $Co(OAc)_2 \cdot 4H_2O$. The new N,S-acetals of acetylketene (XVI-XVIII) were synthesized in this way from compounds (IX-XI).



(IX)—(XI) (XVI)—(XVIII) (2)

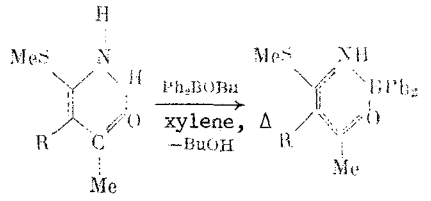
R = Me (IX), (XVI); Et (X), (XVII); *n*-Pr (XI), (XVIII).

It should be noted that the deacylation of N,S-ADK takes place more quickly and with a smaller amount of catalyst than the process in the case of the aminals of diacylketenes [12]. In the preliminary communication [13] we indicated that the transformation (IX) → (XVI) also takes place on boiling over SiO_2 in chloroform, but the yield of compound (XVI) here was somewhat lower than with heating with methanol in the presence of the cobalt salt.

The spectral data of compounds (XVI-XVIII) indicate the presence of intramolecular hydrogen bonds in these compounds. Thus, in addition to the signal for the free NH group at 3490-3485 cm^{-1}, the IR spectra contain absorption bands for the combined NH group in the region of 3300-2850 cm^{-1}.

The ^1H and ^{13}C NMR spectra of compounds (XVI-XVIII) also indicate the presence of only one of the two possible isomeric forms in the solutions of these compounds in deuterochloroform, and this is clearly the E isomer stabilized by an intramolecular (cf. [12]). Thus, the PMR spectrum of compound (XVI) at –55°C contains one set of signals, and the free NH group has a chemical shift of 5.82 ppm, while the NH group involved in the intramolecular hydrogen bond has a chemical shift of 10.28 ppm.

The synthesized N,S-acetals are of interest both as reagents for heterocyclic synthesis and as chelating ligands. Thus, the respective chelate complexes (XIX, XX) were obtained with high yields by the reaction of compound (IX) or (XVI) with Ph_2BOBu.

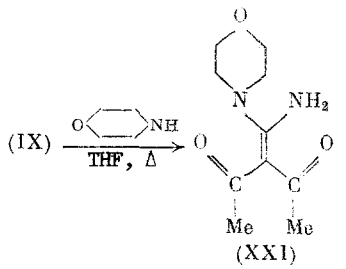


(IX), (XVI) (XIX), (XX) (3)

R = MeCO (IX), (XIX); H (XVI), (XX).

The structure of the crystalline compounds (XIX, XX) was confirmed by spectral methods. In the ^{11}B NMR spectra they give signals in the region of tetracoordinated boron. The mass spectra contain strong peaks for $[M - Ph]^+$ ions.

The N,S-ADK and N,S-AAK can also be used for the synthesis of the aminals of the respective ketenes. For example, the action of morpholine on compound (IX) gave 3-[morpholino(amino)methylene]pentane-2,4-dione (XXI) with a yield of 77%.



(XXI)

The reaction of compound (IX) with aryl isocyanates leads to functionally substituted pyrimidin-2-ones [19].

EXPERIMENTAL

The PMR spectra were recorded on a Bruker WM-250 instrument. The ^{13}C and ^{11}B NMR spectra were obtained on a Bruker AM-300 spectrometer (deuterochloroform, δ, ppm). The IR spectra were recorded on a UR-20 instrument (chloroform, ν, cm^{-1}). The mass spectra (m/z) were obtained on a Varian MAT CH-6 spectrometer.

The initial aryl thiocyanates were obtained by the method in [20] and were purified from aryl isothiocyanate impurities by column chromatography using silica gel with hexane as eluant.

N,S-Acetals of Diacylketenes (IX-XIV). A mixture of 16 mmoles of (I), 6 mmoles of (IV-VIII), and 0.3-0.6 mmole of $Ni(acac)_2$ was stirred under argon for 7-15 days at 20°C. [In the case of compound (XIV) equimolar amounts of (II) and (IV) in THF solution were used.] The process was monitored by IR spectroscopy from the disappearance of the absorption band of the C≡N group. The excess of compound (I) or solvent was distilled under vacuum, and the residue was crystallized from a mixture of benzene and hexane. [An alternative method for the purification of compounds (IX-XI) involves sublimation of the crude products at 60-80°C (1 mm Hg).]

3-[(Methylthio)aminomethylene]pentane-2,4-dione (IX). The yield was 72%; mp 118-119°C (1:1 benzene-hexane). Mass spectrum: 173 $[M]^+$. Found: C 48.75; H 6.45; N 8.22; S 18.63%. $C_7H_{11}NO_2S$. Calculated: C 48.53; H 6.40; N 8.09; S 18.51%. IR spectrum: 3490 (NH), 3230-2820 (NH, CH), 1585 (CO, C=C). PMR spectrum (-30°C): 12.26 br.s (NH), 7.03 br.s (NH), 2.32 s and 2.39 s (3Me). (At ~20°C the signals of the NH_2 group are not observed in the spectrum on account of strong broadening.) ^{13}C NMR spectrum: 196.73 (2 CO), 172.37 (SCN), 113.51 (C^3), 31.30 (2 Me CO), 13.56 (Me).

3-[(Ethylthio)aminomethylene]pentane-2,4-dione (X). The yield was 61%; mp 72-74°C (4:5 benzene-hexane). Mass spectrum: 187 $[M]^+$. Found: C 50.87; H 7.15; N 7.60; S 16.98%. $C_8H_{13}NO_2S$. Calculated: C 51.31; H 7.00; N 7.48; S 17.12%. IR spectrum: 3500 (NH), 3220-2850 (NH, CH), 1590, 1550 (CO, C=C). PMR spectrum: 2.80 q (CH_2), 2.40 s (2Me), 1.40 t (Me).

3-[(Propylthio)aminomethylene]pentane-2,4-dione (XI). The yield was 79%; mp 68-69°C (1:1 ether-hexane). Mass spectrum: 201 $[M]^+$. Found: C 53.83; H 7.43; N 7.11; S 15.84%. $C_9H_{15}NO_2S$. Calculated: C 53.70; H 7.51; N 6.96; S 15.93%. IR spectrum: 3500 (NH), 3200-2850 (NH, CH), 1590, 1540 (CO, C=C). PMR spectrum: 2.76 t (CH_2S), 2.37 s (2Me), 1.95-1.53 m (CH_2), 1.05 t (Me).

3-[(Phenylthio)aminomethylene]pentane-2,4-dione (XII). The yield was 75%; mp 112-113°C (1:2 benzene-hexane). Mass spectrum: 253 $[M]^+$. Found: C 61.43; H 6.05; N 6.15; S 13.49%. $C_{12}H_{13}NO_2S$. Calculated: C 61.25; H 5.57; N 5.95; S 13.63%. IR spectrum: 3440 (NH), 3200-2840 (NH, CH), 1635, 1585, 1577, 1565 (CO, C=C). PMR spectrum: 7.57-7.38 m (Ph), 2.37 s (2Me). ^{13}C NMR spectrum: 196.82 (2CO); 171.46 (SCN); 136.17; 130.97; 130.31; 127.26 (Ph); 112.52 (C^3); 31.45 (2Me).

3-[(p-Tolylthio)aminomethylene]pentane-2,4-dione (XIII). The yield was 87%; mp 123-125°C (1:2 benzene-hexane). Mass spectrum: 249 $[M]^+$. Found: C 62.59; H 6.21; N 6.13; S 12.72%. $C_{13}H_{15}NO_2S$. Calculated: C 62.62, H 6.06, N 5.62, S 12.86%. IR spectrum: 3435 (NH), 3200-2900 (NH, CH), 1630, 1590, 1540 (CO, C=C). PMR spectrum: 7.40 d (2H, Ph), 7.25 d (2H, Ph), 2.38 s (2MeCO), 2.33 s (Me). ^{13}C NMR spectrum: 196.90 (2CO); 172.17 (SCN); 141.77; 136.27; 131.25; 123.65 (Ph); 112.51 (C^3); 31.54 (2MeCO); 21.32 (Me).

2-[(Methylthio)aminomethylene]-1-phenylbutane-1,3-dione (XIV). The yield was 66%; mp 101-102°C (hexane). Mass spectrum: 235 $[M]^+$. Found: C 61.44; H 5.73; N 5.75; S 13.75%. $C_{12}H_{13}NO_2S$. Calculated: C 61.25; H 5.57; N 5.95; S 13.63%. IR spectrum: 3450 (NH); 3300-2900 (NH, CH); 1600; 1590; 1550 (CO, C=C). PMR spectrum: 11.90 br.s (NH), 6.07 br.s (NH), 7.85-7.35 m (Ph), 2.34 s (MeCO), 1.18 s (Me).

Compound (XIV) was also synthesized with 0.5 mmole of $Ni(bac)_2$ instead of $Ni(acac)_2$. In this case the reaction time was 12 days, and the yield of (XIV) was 77%.

Ethyl 2-[(Methylthio)aminomethylene]-3-oxobutyrate (XV). The compound was obtained similarly to compounds (IX-XIII) from acetoacetic ester (III) and methyl thiocyanate (IV). The yield was 40%; mp 64-65°C (1:1 benzene-hexane). Mass spectrum: 203 $[M]^+$. Found %: C 47.13; H 6.67; N 7.00; S 15.78%. $C_8H_{13}NO_2S$. Calculated %: C 47.27; H 6.45; N 6.89; S 15.78%. IR spectrum: 3500 (NH), 3200-2850 (NH, CH), 1670 (CO_2Et), 1600, 1570 (CO, C=C). PMR spectrum: 12.60 br.s (NH), 6.00 br.s (NH), 4.20 q (CH_2), 2.32 s, 2.27 s (2Me), 1.28 t ($MeCH_2$) ^{13}C NMR spectrum: 195.72 (COMe), 174.04 (CO), 168.74 (SCN), 101.16 (C^2), 60.02 (CH_2), 31.11 (MeCO), 14.21 ($MeCH_2$), 13.53 (Me).

N,S-Acetals of Acetylketene (XVI-XVIII). A solution of 10 mmoles of the N,S-ADK (IX-XII) and 0.2 mmole of $Co(OAc)_2 \cdot 4H_2O$ in 20 ml of methanol was boiled for 2.5-3 h (monitored by TLC). The solvent was distilled under vacuum, and the residue was chromatographed on a short column of silica gel. The colored impurity was eluted with benzene, and the products (XVI-XVIII) were then eluted with chloroform.

1-Amino-1-methylthiobut-1-en-3-one (XVI). The yield was 84%; mp 109-110°C (1:1 benzene-hexane). Mass spectrum: 131 $[M]^+$. Found: C 46.05; H 7.00; N 10.83; S 24.02%. C_5H_9NOS. Calculated: C 45.77; H 6.91; N 10.68; S 24.44%. IR spectrum: 3485 (NH), 3270-2830 (NH, CH), 1612, 1580 (CO, C=C). PMR spectrum (-50°C): 10.28 br.s, (NH), 5.82 br.s (NH), 5.10 s (CH=), 2.32 s, 1.98 (2Me). ^{13}C NMR spectrum: 193.77 (CO), 165.17 (SCN), 92.34 (C^2), 29.04 (MeCO), 13.12 (Me).

1-Amino-1-ethylthiobut-1-en-3-one (XVII). The yield was 87%; mp 56-57°C (hexane, 10°C). Mass spectrum: 145 $[M]^+$. Found: C 49.91; H 7.84; N 9.24; S 22.04%. $C_6H_{11}NOS$. Calculated: C 49.62; H 7.63; N 9.65; S 22.08%. IR spectrum: 3490 (NH), 3300-2850 (NH, CH), 1610, 1580 (CO, C=C). PMR spectrum: 5.12 s (CH=), 2.88 q (CH_2), 2.05 s (MeCO), 1.34 t (Me).

1-Amino-1-propylthiobut-1-en-3-one (XVIII). The yield was 89%; mp 49-50°C (ether, -70°C). Mass spectrum: 159 $[M]^+$. Found: C 52.90; H 8.28; N 8.95; S 19.89%. $C_7H_{13}NOS$. Calculated: C 52.79; H 8.23; N 8.80; S 20.13%. IR spectrum: 3490 (NH), 3300-2850 (NH, CH), 1613, 1570 (CO, C=C). PMR spectrum: 5.14 s (CH=), 2.78 t (CH_2S), 1.98 s (MeCO), 1.88-1.42 m (CH_2), 0.99 (Me).

Synthesis of the Diphenylboryl Chelate (XIX) from (IX) and Ph_2BOBu. A mixture of 10 mmoles of (IX) and 20 mmoles of Ph_2BOBu in 30 ml of xylene was boiled for 3 h. The solvent was distilled, the residue was washed with hexane, and 2.60 g (77%) of the chelate (XIV) was obtained; mp 130-131°C (1:2 benzene-hexane). Mass spectrum: 260 $[M - Ph]^+$. Found: C 68.05; H 5.74; B 3.15; N 4.06; S 9.45%. $C_{19}H_{20}BNO_2S$. Calculated: C 67.66; H 5.98; B 3.21; N 4.15; S 9.51%. IR spectrum: 3403 (NH), 1660 (CO), 1567, 1552 (CO, C=C). PMR spectrum: 7.46-7.23 m (2Ph), 7.01 br.s (NH), 2.51 s, 2.36 s, 2.32 s (3Me). ^{13}C NMR spectrum: 196.86 (CO), 182.89 (CO → B), 173.43 (SCN), 149.50, 131.80, 127.41, 126.88 (2Ph), 114.59 (C^3), 31.87, 25.41 (2 Me), 13.50 (MeS). ^{11}B NMR spectrum: 2.8.

Synthesis of the Diphenylboryl Chelate (XX) from (XVI). Compound (XX) was obtained by analogy with the previous method, and the reaction time was 2 h. The yield was 81%; mp 119-120°C (1:1 benzene-hexane). Mass spectrum: 218 $[M - Ph]^+$. Found: C 69.60; H 6.13; B 3.74; N 4.80; S 10.67%. $C_{17}H_{18}BNOS$. Calculated: C 69.16; H 6.15; B 3.67; N 4.75; S 10.86%. IR spectrum: 3410 (NH), 1585, 1520, (CO, C=C). PMR spectrum: 7.54-7.28 m (2 Ph), 6.60 br.s (NH), 5.38 s (CH=), 2.47 s, 2.20 s (2Me). ^{13}C NMR spectrum: 177.11 (CO), 172.10 (SCN), 150.0, 131.86, 127.31, 126.37 (2Ph), 95.01 (C^2), 23.61 (MeCO), 12.30 (MeS). ^{11}B NMR spectrum: 2.1.

3-[Morpholino(amino)methylene]pentane-2,4-dione (XXI). A solution of 10 mmoles of compound (IX) and 11 mmoles of morpholine in 20 ml of THF was boiled for 5 h. The crystals which separated on cooling were filtered off, and 1.63 g (77%) of compound (XXI) was obtained; mp 207-208°C (benzene). Mass spectrum: 212 $[M]^+$. Found: C 56.76; H 7.45; N 13.07. $C_{10}H_{16}N_2O_3$. Calculated: C 56.59; H 7.60; N 13.20. IR spectrum (tablets with potassium bromide): 3400-2800 (NH, CH), 1685 (CO), 1585, 1560 (C=C). PMR spectrum (DMSO-d_6): 8.57 br.s (NH), 8.17 br.s (NH), 3.64 t ($2CH_2$), 3.48 t (2 CH_2), 1.97 s (2Me).

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