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INVESTIGATION OF THE REGIO- AND STEREOSELECTIVITY OF THE ANNELATION REACTION OF CYCLIC SCHIFF BASE WITH STRUCTURALLY ASYMMETRICAL β,β'-TRIKETONES. SYNTHESIS AND PROPERTIES OF 17-ACETOXY-8-AZA-D-HOMOGONA-12,17a-DIONES

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Annelation ([2+4]-cyclocondensation) of 3,4-dihydroisoquinolines and 2-acetyl-4-acetoxycyclohexane-1,3-dione gives 17-acetoxy-8-aza-D-homogona-12,17a-diones as a mixture of the C(9),C(17)-stereoisomers with the (9R,17S: 9S,17R) racemic pair predominating.

Keywords: 8-aza-D-homogonanes, 2-acylcyclohexane-1,3-diones, 3,4-dihydroisoquinoline, annelation, regiospecificity, stereochemistry, β,β'-triketone tautomerism

In examples of the cyclocondensation of 3,4-dihydroisoquinolines with structurally asymmetrical 2-acetylcyclohexane-1,3-dione derivatives, a high degree of regio- and stereoselectivity was identified for the annelation reaction of cyclic Schiff bases with structurally asymmetrical β,β'-triketones [1-3] and some limits for the use of this reaction have been determined [4-6]. The results obtained are of considerable theoretical and practical interest for investigating the mechanism of such reactions in the development of routes to the synthesis of 8-aza steroids with specified regio- and stereochemistry. However, not all aspects of the regio- and stereoselectivity of these reactions were clear. In particular, it was not clear whether the discovered regio- and stereoselectivity was dictated by the presence of a substituent at the C(4) atom of 2-acyl-5,5-dimethylcyclohexane-1,3-diones or whether the presence of the C(5) *gem*-dimethyl group was also important and also what kind of role each of these factors plays in the achievement of the regio- and stereochemical result. With the aim of elucidating these questions we began an investigation of the indicated reaction as applied to the recently available 2-acyl-4-acyloxycyclohexane-1,3-diones [7].

Condensation of 3,4-dihydroisoquinolines (**1a,b**) with 2-acetyl-4-acetoxycyclohexane-1,3-dione (**2**) was brought about by refluxing equimolar mixtures of reagents or by holding them at room temperature, in agreement with the conditions given in [1-3]. Both in the first and in the second case the tetracyclic 8-aza-D-homogonane derivative target products (**3a,b**) were produced.

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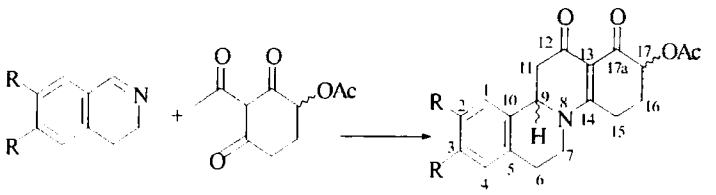
Scheme 1



1a,b **2** **3a,b**

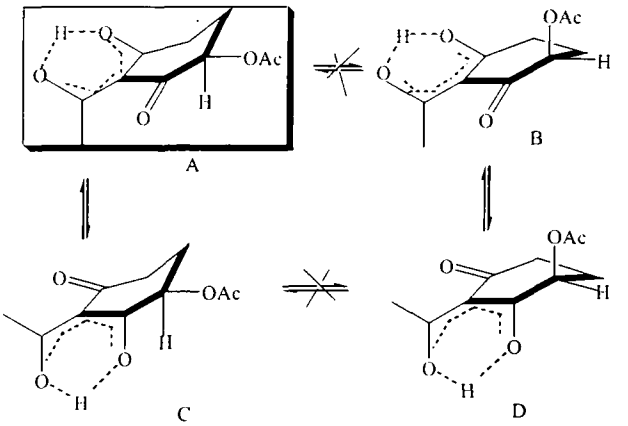
a R = H, **b** R = OMe

According to TLC data, 8-aza-D-homogonanes **3a,b** were obtained as a chromatographically poorly differentiated mixture of two components. Attempts to separate these mixtures by chromatography or by fractional crystallization proved unsuccessful.

Bearing in mind the racemic character of β,β'-triketone **2**, the formation of a new chiral center at C(9) in the course of the reaction, and also the previously indicated regioselectivity of such cyclocondensations [1-3], it could be affirmed that we had obtained a mixture of four C(9)-, C(17)-stereoisomers. Moreover, the racemic pair (9R,17S: 9S,17R) and (9R,17R: 9S,17S) of diastereomers gave two spots on TLC.

With the aim of revealing and explaining the stereochemical results for this reaction we considered the spatial structure of 2-acyl-4-acetoxycyclohexane-1,3-dione **2** and 3,4-dihydroisoquinolines **1a,b** and the possible transition state for the reaction.

Scheme 2



Analysis of Dreiding molecular models has shown that the cyclohexane rings of 2-acylcyclohexane-1,3-diones and 2-acetyl-4-acetoxycyclohexane-1,3-dione **2** are specifically flattened as a result of the full enolization of the β,β'-tricarbonyl fragment and can achieve two mutually interconverting chair conformers with equatorial (A, C) or axial (B, D) positioning of the C(4)-acetoxy substituent (Scheme 2, shown only for the R-isomer). In addition, the interconversion of the isomers with an equatorial and axial C(4) substitutent is evidently inhibited. A study using ^{1}H NMR spectroscopy has shown that β,β'-triketone **2** is fully enolized and exists in solutions as the individual enol tautomer A. Any additional signals, which might be assigned to tautomer C as well as signals for a hypothetical conformer with an axially placed substituent B or D, were not seen in the ^{1}H NMR spectra. From this data we conclude that conformers B and D either are not realized or are present in very low concentration. This may be a result of stereoelectronic repulsion between an axial 4-acetoxy substitutent and the π-electron cloud of the enolized β,β'-tricarbonyl fragment and hence forms B and D govern neither the regio- nor the stereochemical result of the reaction.

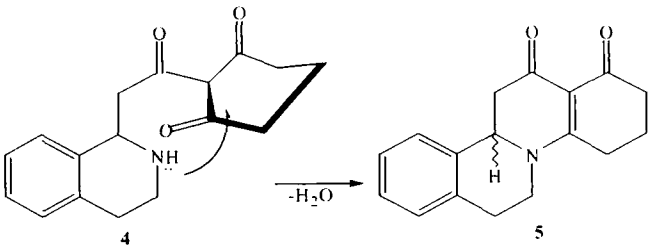
Analysis of the stereostructure of 3,4-dihydroisoquinolines **1a,b**, in turn, showed that the dihydropyridine ring of 3,4-dihydroisoquinolines exists as a rapidly interconverting chair conformer. This conclusion is a consequence of the nature of the observed resonance signals for the protons at C(3) and C(4) of 3,4-dihydroisoquinolines which appear as A_2X_2 type individual triplets [8], being observed at 2.60-2.70 (C(4)) and 3.60-3.70 (C(3)) ppm (J = 7-8 Hz), and shifted to low field at 3.00-3.30 and 3.90-4.20 ppm (J = 8.0-8.5 Hz) respectively upon quaternization (protonation).

Scheme 3

From this it follows that attack of the symmetrical β,β'-triketone by the azomethine fragment of 3,4-dihydroisoquinoline is equally likely from the front and from the back.

It has previously been proposed that cyclocondensation of 3,4-dihydroisoquinolines with 2-acylcycloalkane-1,3-diones occurs via an alkylation stage with the formation of a tricyclic Mannich base type intermediate which, as a result of subsequent cyclodehydration, is converted to the final 8-aza steroidal ABCD tetracycle **5** [9].

Scheme 4



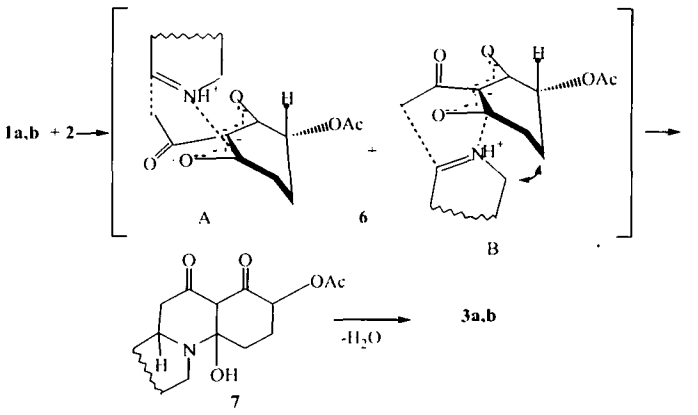
4 5

However, such a sequence of reactions seems unlikely since the geometry of the tricyclic Mannich base does not fit in with the stereochemical requirements of attack by the nitrogen atom on the carbonyl groups of the cycloalkane-1,3-dione fragment, as suggested by the Dreiding models. Additionally, not in one of the previously reported investigations [1-6], even when applied to sterically hindered 3,4-didhydroisoquinolines and 2-acylcycloalkane-1,3-diones [4-6], were such adducts observed. Even so, if one assumes formation of adducts **4**, one would expect them to be extremely unstable and to undergo a retro-Michael type fission to the starting materials under the reaction conditions. With this under consideration and bearing in mind the previously mentioned regio- and steroselectivity of the discussed reaction (together with the case that for noncyclic Schiff bases, having in contrast to the cyclic bases a *trans* configuration of the azomethine fragment, such reactions are not observed) it might be expected that the discussed reactions occur by a coordinated mechanism with simultaneous formation of the C–C and C–N bonds.

2-Acylcycloalkane-1,3-diones are CH-acids (pK_a 7-10) (DMF) [10] and 3,4-dihydroisoquinolines are bases [11] hence they can form salts with one another which, in some cases, have been separated and characterized [6, 9] but are still little studied. It was found that formation of salts of 3,4-dihydroisoquinolines with 2-acylcyclohexane-1,3-diones have not been successfully recorded [1, 12] but, none the less, this does not exclude their formation and participation in the discussed reactions.

Hence our overall position is to propose the following reaction scheme for annelation of 3,4-dihydroisoquinolines **1a,b** with 2-acetyl-4-acetoxycyclohexane-1,3-dione **2**. As a result of the interaction of the quasi-basic 3,4-dihydroisoquinolines with the quasi-acidic β,β'-triketone there are formed salts which, *via* a six membered transition state (**6A**- attack by 3,4-dihydroisoquinoline from the front and **6B**- attack from the back as shown for the R-isomer of β,β'-triketone), are converted to the hypothetical alcohol **7**. Dehydration of the latter under these reaction conditions completes the conversion and gives the final 8-aza-D-homogonanes **3a,b** as a mixture of stereoisomers.

Scheme 5



Analysis of the transition states **6A** and **6B** on Dreiding models has shown that attack from the back is less preferred due to steric hindrances arising from the steric interactions of the C(3)- and C(5)-methylene groups of 3,4-dihydroisoquinolines and β,β'-triketone respectively. It can therefore be concluded that, as a result of this reaction, there are preferentially formed racemates with (9*R*,17*S*:9*S*,17*R*), and not (9*R*,17*R*:9*S*,17*S*) configurations.

The structure assigned to 17-acetoxy-8-aza-D-homogona-12,17a-diones **3a,b** was confirmed by physicochemical data. 17-Acetate of 8-aza-D-homogonane **3a** was obtained as a crystallohydrate with one equivalent of water and this was confirmed by elemental analysis and by IR spectroscopy. Dehydration of the crystallohydrate in vacuo over P_2O_5 at 110°C gave yellow crystals with mp 170-180°C (with decomp.) and showed the absence in the IR spectrum of a broad absorption band at 3600-3300 cm^{-1} for the hydrogen bound OH groups. TLC of this sample showed partial decomposition. Recrystallization from a mixture of absolute alcohol and ether gave a colorless, chromatographically pure sample which, according to ^1H NMR data, is a crystallosolvate with 0.5 equivalents of EtOH and has mp 129-131°C. These results point to a specific tendency for 8-aza-D-homogonane **3a** to crystallize as a crystallosolvate. 17-Acetate of 8-aza-D-homogonane **3b** crystallizes without the inclusion of solvent in the crystal lattice. For the condensation products **3a,b** their IR spectra show absorption bands at ~1740 and 1250-1230 cm^{-1} corresponding to the 17-acetoxy substituent, and in the region 1700-1400 cm^{-1} which are typical of N(8)–C(14)=C(13)–[C(12)=O]–C(17a)=O in an aminovinyldicarbonyl fragment [12, 13]. The electronic absorption spectra of derivatives **3a,b** show two broad absorption bands at ~270 and ~300 nm, corresponding to the electronic transition of the aminovinyldicarbonyl fragment [12, 14] and, for derivative **3b**, also at 233 nm due to the absorption of the dimethoxy substituted aromatic ring A.

Confirmation of the ABCD-tetracyclic structure assigned to the derivatives **3a,b** followed from the nature of the resonance absorption signals in their ^1H NMR spectra. Hence, in the spectra of acetates **3a,b**, there are observed singlet resonance absorptions for the methyl groups of the 17-acetoxy substituent in the region 2.16-2.18 ppm and a double doublet at 5.15-5.16 ppm for the quasi-axially placed proton on C(17). The above data proves that the acetoxy substituent in the 8-aza-D-homogonane molecules **3a,b** are positioned on carbon atom C(17) and not on C(15). In the latter case, the equatorial placement of the acetoxy group would experience a

marked steric interaction with C(7) of the methylene groups. It was noted that, in the ^{1}H NMR spectrum of **3a**, additional signals corresponding to a second racemic pair were absent. For compound **3b** there are present additional signals for some protons groups, in particular for C(2)OMe, C(3)OMe, C(1)H, C(4)H, C(9)H, and C(17)OCOCH$_3$ and their integrated intensities allowed one to infer that racemates were present in a 2:1 ratio. This then confirms the proposal regarding the preference of transition state **6A** when compared with transition state **6B**.

Convincing evidence regarding formation of two diastereomers in compounds **3a,b** comes from the ^{13}C NMR spectrum of acetate **3b** in which almost all of the ^{13}C absorption resonances have companion signals of low intensity (data in the Experimental given in square brackets) due to the presence of the second racemic pair. The number of basic ^{13}C NMR resonance signals (21 peaks) for acetate **3b** corresponds to three primary, five secondary, four tertiary, and nine quaternary ^{13}C carbon nuclei and are found in the expected regions of the spectrum thus confirming the structure assigned to compound **3b**.

Hence the above data confirms that the (9*R*,17*S*:9*S*,17*R*) racemate of derivative **3b** predominates and does, in fact, fit the ^{1}H and ^{13}C NMR spectra given.

We therefore conclude that annelation of 3,4-dihydroisoquinolines **1a,b** by 2-acetyl-4-acetoxycyclohexane-1,3-dione **2** occurs regioselectively with the preferred formation of the (9*R*,17*S*:9*S*,17*R*) racemic pair of 17-acetates of 8-aza-D-homogonanes **3a,b**. Evidently, the use of an enantiomer of β,β'-triketone **2** (e.g. with the (*R*)-configuration) will lead to the formation of a mixture of only two (9*R*,17*R*)- and (9*S*,17*R*)- diastereomers and that these, in principle, may be separated into individual, optically active diastereomers. As a result, this opens a route to the preparation of optically active 8-aza steroids with a desired stereochemistry. We also propose that the use of enantiomers of 4-substituted derivatives of 2-acyldimedone (for which the transition state **6B** cannot be achieved due to the steric hindrance caused by the C(5)-*gem*-dimethyl grouping) can lead to a stereoselective reaction giving 8-aza steroids with a defined stereochemistry at the C(9) chiral center, the configuration of which is determined by the configuration of the chiral β,β'-triketone.

EXPERIMENTAL

3,4-Dihydroisoquinolines **1a,b** used in this work were obtained by cyclodehydration of phenethylformamides using polyphosphoric acid (azomethine **1a**) or phosphorus oxychloride (azomethine **1b**) under Bischler–Napieralski reaction conditions [15]. Monitoring of the reaction course was carried out by TLC on Silicagel F_{60} 254 plates using chloroform–methanol (9:1) eluent. Melting points were measured on a Boetius heating block. IR Spectra were obtained on a UR-20 instrument for KBr tablets. UV Spectra were taken on a Specord M-400 spectrophotometer using methanol. Mass spectra were obtained on a Shimadzu MS QP-5000 spectrometer with direct introduction of the sample and an electron ionization energy of 70 eV. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker AC-200 radiospectrometer with working frequencies of 200 MHz for protons and 90.53 MHz for carbon using $CDCl_3$ solvent and TMS as internal standard.

9ξ,17ξ-Acetoxy-8-aza-D-homogona-1,3,5(10),13-tetraene-12,17a-dione (3a). A mixture of 3,4-dihydroisoquinoline **1a** (0.131 g, 1.0 mmol) and β,β'-triketone **2** (0.212 g, 1.0 mmol) in ethanol (5 ml) was allowed to stand at room temperature, following the course of the reaction by TLC. After 12 h, a crystalline phase began to separate from the reaction mixture. After 36 h, following the disappearance of the starting material, the reaction mixture was diluted with ether to turbidity and then left at 5°C for 12 h for completion of the crystallization. The material produced was filtered off and recrystallized from a mixture of alcohol and ether (1:3) to give the crystallohydrate of 8-aza-D-homogonane **3a** as colorless, finely needled crystals. Yield 93%; mp 127-130°C. IR spectrum, ν, cm^{-1}: 3600-3300, 3000-2800, 1745, 1691, 1626, 1595 sh, 1535, 1503, 1468, 1420, 1397, 1362, 1327, 1262-1232, 1150, 1074, 892, 782, 763. UV spectrum, λ_{max}, nm (log ε): 265 (4.17), 303.9 (4.26); λ_{min} nm (log ε): 280 (3.98). ^{1}H NMR spectrum, δ, ppm, *J* (Hz): 1.70-2.40 (2H, m, C(16)H$_2$); 2.16 (3H, s, OCOCH$_3$); 2.57 (1H, t, C(11)H$_B$, *J* = 15.0); 2.82 (1H, dd, C(11)H$_A$, *J* = 3.0, *J* = 15.0); 2.86-3.26 (4H, m, C(6)H$_e$, C(6)H$_a$, C(15)H$_2$); 3.46 (1H, m, C(7)H$_a$); 4.18 (1H, m, C(7)H$_e$); 5.00 (1H, dd, C(9)H$_X$, *J* = 3.0, *J* = 15.0); 5.15 (1H, dd, C(17)H, *J* = 4.5, *J* = 13.0); 7.08-7.42 (4H, m, C(1)H, C(2)H, C(3)H, C(4)H). Found, %: C 66.40; H 6.09; N 3.97. [M-18]$^+$ 325. $C_{19}H_{19}NO_4 \cdot H_2O$. Calculated, %: C 66.46, H 6.16, N 4.08. M 343.38.

Anhydrous **3a**, mp 170-180°C (decomp.). IR spectrum, ν, cm^{-1}: 3000-2830, 1745 1690, 1629, 1600 sh, 1540, 1505, 1465-1455, 1420, 1395, 1360, 1330, 1265-1230, 1150, 1075, 890, 785, 761.

Crystallosolvate of **3a** with 0.5 equivalents of EtOH, colorless crystals; mp 129-131°C. ^{1}H NMR spectrum, δ, ppm, *J* (Hz): 1.25 (1.5 H, t, *J* = 7.0, CH_3 (EtOH)); 1.85-2.40 (2H, m, C(16)H_2); 2.18 (3H, s, $OCOCH_3$); 2.55 (1H, t, *J* = 15.5, C(11)H_B); 2.81 (1H, dd, *J* = 4.0, *J* = 15.5, C(11)H_A); 2.85-3.25 (4H, m, C(6)H_a, C(6)H_e, C(15)H_2); 3.43 (1H, dtd, *J* = 4.0, *J* = 12.0, *J* = 14.0, C(7)H_a); 3.72 (1H, q, *J* = 7.0, CH_2 (EtOH)); 4.20 (1H, tt, *J* = 4.0, *J* = 14.0, C(7)H_e); 5.05 (1H, dd, *J* = 4.0, *J* = 15.5, C(9)H_X); 5.16 (1H, dd, *J* = 4.0, *J* = 12.0, C(17)H); 7.02-7.36 (4H, m, C(1)H, C(2H), C(3)H, C(4)H).

9ξ,17ξ-Acetoxy-2,3-dimethoxy-8-aza-D-homogona-1,3,5(10),13-tetraene-12,17a-dione (3b). A mixture of 3,4-dihydroisoquinoline **1b** (0.191 g, 1.0 mmol) and β,β'-triketone **2** (0.212 g, 1.0 mmol) in ethanol (7 ml) was refluxed under an argon atmosphere, following the course of the reaction by TLC. After 3 h the reaction mixture was evaporated by a half, diluted with ether to turbidity, and left in the cold. The precipitated crystals were filtered off, washed with ether, and recrystallized from a mixture of alcohol and ether (1: 4) to give 8-aza-D-homogonane **3b** as pale yellow, finely needled crystals. Yield 88%; mp 206-207.5°C (decomp.). IR Spectrum, ν, cm^{-1}: 3000-2830, 1742, 1690, 1621, 1550-1500, 1461, 1419, 1377 sh, 1344, 1325, 1263, 1252-1229, 1210 sh, 1188, 1139, 1072, 1030, 1000, 893, 869, 811, 775. UV spectrum, λ_{max}, nm (log ε): 233.2 (3.88), 267.3 (4.17), 301.8 (4.25). λ_{min}, nm (log ε): 217.8 (3.60), 245 (3.69), 278.2 (4.11). ^{1}H NMR spectrum, δ, ppm, *J* (Hz): 1.90-2.38 (2H, m, C(16)H_2); 2.18 (3H, s, $OCOCH_3$); 2.54 (1H, t, *J* = 15.5, C(11)H_B); 2.82 (1H, dd, *J* = 4.0, *J* = 15.5, C(11)H_A); 2.84-3.20 (4H, m, C(6)H_a, C(6)H_e, C(15)H_2); 3.42 (1H, ddd, *J* = 3.0, *J* = 12.0, *J* = 12.0, C(7)H_a); 3.89 (6H, s, C(2)OCH_3), C(3)OCH_3); 4.22 (1H, tt, *J* = 3.0, *J* = 3.0, *J* = 12.0, C(7)H_e); 4.90 (1H dd, *J* = 4.0, *J* = 15.5, C(9)H_X); 5.16 (1H, dd, *J* = 5.5, *J* = 13.0 C(17H); 6.68 (2H, s, C(1)H, C(4)H). Additional signals with 0.5 the intensity of the primary signal: 2.14 s ($OCOCH_3$); 2.60 (t, *J* = 15.0, C(11)H_B); 2.81 (dd, *J* = 5.0, *J* = 15.0, C(11)H_A); 3.86 s and 3.87 s C(2)OCH_3 and C(3)OCH_3; 4.82 (dd, *J* = 5.0, *J* = 15.0, C(9)H_X); 5.24 (dd, *J* = 5.0, *J* = 10.5, C(17)H); 6.62 s and 6.66 s C(1)H, C(4)H. ^{13}C NMR spectrum, δ, ppm: 29.954 ($OCO\underline{C}H_3$), 26.011 [24.883] (CH_2), 26.261 [27.166] (CH_2), 29.522 [29.142] (CH_2), 44.871 [45.193] (CH_2), 46.239 [45.689] (CH_2), 56.025 (OMe), 56.145 (OMe), 56.143 [57.397] (C(9)), 65.509 [65.231] (C(17)), 102.164 [104.025] (C(13)), 108.650 [108.793] (C(4)), 111.022 [110.899] (C(1)), 125.162 [125.398] (C(10)), 125.309 [126.678] (C(5)), 148.500 [148.398] (C(3)), 148.622 (C(2)), 169.327 [169.451] ($O\underline{C}OCH_3$), 170.243 [170.480] (C(14)), 186.209 [185.451] (C=O), 188.543 [189.957] (C=O). Found, %: C 65.34; H 5.96, N 3.55. [M]$^+$ 385. $C_{21}H_{23}NO_6$. Calculated, %: C 65.44, H 6.01, N 3.63. M 385.42.

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