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PREPARATION OF cis AND trans-CYCLOALKANE-CONDENSED PYRIMIDINE-DIONES BY AZETIDINONE RING TRANSFORMATION

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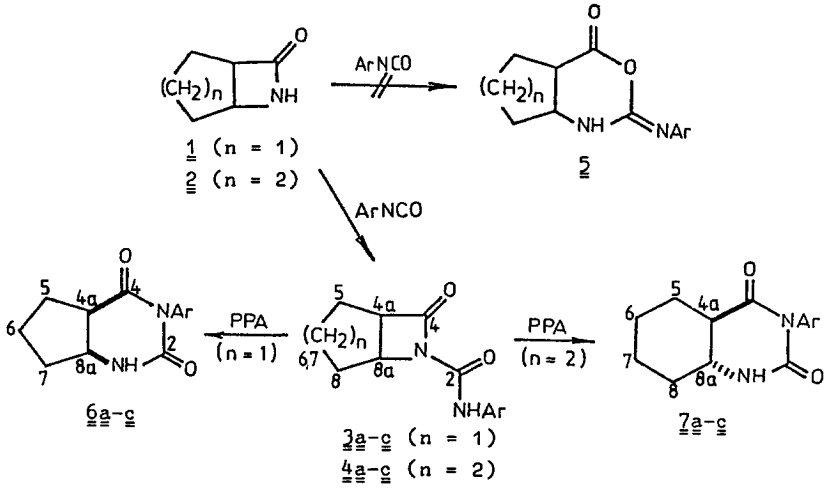
Abstract - N-substituted derivatives (3 and 4) obtained from cyclopentane- and cyclohexane-azetidinones (1 and 2) were isomerized with polyphosphoric acid to give cyclopentane-cis- (6) and cyclohexane-trans-condensed (7) 2,4-pyrimidinediones. The structures of the dihydrouracils prepared by the ring transformation were proved by ^{1}H- and ^{13}C-nmr spectroscopy and by comparison with the compounds synthesized from cis- and trans-2-amino-1-cycloalkane-carboxamides (11-13) with 1,1'-carbonyldiimidazole.

Earlier, the diexo-norbornane and -norbornene-condensed azetidinones were transformed with polyphosphoric acid (PPA) to methylene-bridged quinazoline-diones, and from derivatives containing a double bond in the bicycle 3-substituted uracils were obtained by thermal cycloreversion.[2] This new method is now applied for the synthesis of non-methylene-bridged cycloalkane-condensed pyrimidinediones. The ring enlargement of the strained azetidinone for the preparation of different heterocycles is an obvious and often applied synthetic method.[3-9]

RESULTS

Cycloalkane-cis-condensed azetidinones (1 and 2) were prepared from cyclopentene and cyclohexene through the addition of chlorosulphonyl isocyanate and subsequent reduction. With aryl isocyanates, the azetidinones 1 and 2 furnished the urea derivatives 3 and 4 (Scheme 1). A comparison of the ir, ^{1}H- and ^{13}C-nmr data (Tables 1 and 2) with those on the nor-

bornane/ene analogues investigated earlier[2] permitted exclusion of the possible imino-1,3-oxazinone structure 5.



a: Ar = C_6H_5; b: Ar = C_6H_4Cl(m); c: Ar = C_6H_4Cl(p)

Scheme 1

Table 1. Ir and ^{1}H-nmr data on compounds 3a-c, 6a-c, 7a-c and 14a-c[a]

Compound	νNH band	νC=O band	CH_2 groups m' s (6 or 8H)	H-4a m[b](1H)	H-8a m[c](1H)	NH s(1H)	ArH m(5/4H)[d]
3a	3330	1757 1705	1.4 - 2.5	3.59	4.52	8.55	7.05[e] 7.30[f] 7.55
3b	3295	1761 1688	1.4 - 2.5	2.63	4.54	8.55	7.06[e] 7.25[g] 7.62[h]
3c	3320	1751 1705	1.4 - 2.05	3.62	4.55	8.55	7.27[j] 7.43[f]
6a	3235	1726 1684	~1.8[i] ~2.05[g]	2.92	3.92	7.80	~7.15[g,j] ~7.4[k]
6b	3285	1730 1684	~1.8[i] ~2.05[g]	2.93	3.92	7.90	7.15[l] 7.32[h] 7.42[g]
6c	3220	1724 1686	~1.8[i] ~2.05[g]	2.93	3.92	7.85	7.20[j] 7.45[f]
7a	3220	1725 1675	~1.25[i] ~1.75[g] ~2.05[g]	2.45	~3.35	8.00	7.12[g,j] ~7.35[k]
7b	3220	1720 1675	~1.25[i] ~1.75[g] ~2.05[g]	2.45	~3.35	8.15	7.15[l] 7.30[h] ~7.40[g]
7c	3225	1730 1688	~1.25[i] ~1.75[g] ~2.05[g]	2.45	~3.35	8.10	7.18[j] 7.45[f]
14a	3235	1725 1685	~1.35[k] ~1.65[i] ~2.05	2.95	~3.6	8.08	7.15[g,j] 7.42[k]
14b	3220	1735 1687	~1.35[k] ~1.65[i] ~2.05	2.95	~3.6	8.45	7.15[l] 7.35[h] 7.45[g]
14c	3255	1726 1687	~1.35[k] ~1.65[i] ~2.05	2.93	~3.6	8.12	7.19[j] 7.48[f]

[a] Ir in KBr, cm^{-1}; ^{1}H-nmr: solvent: DMSO-d_6; $CDCl_3$ for compounds 3a-c, δ_{TMS}: 0 ppm; [b] for 3a-c dd (J: 7.9 and 4.0 Hz), qa for 6a-c (half signal-width $\Delta\nu \approx 25$ Hz) and 14a-c ($\Delta\nu \approx 10$ Hz), ~t for 7a-c (J ≈ 10 Hz); [c] ~t for 3a-c (J ≈ 4.4 Hz), m for 6a-c ($\Delta\nu \approx 20$ Hz), and 14a-c ($\Delta\nu \approx 20$ Hz), hidden by the water signal of the solvent in the case of 7a-c; [d] 2 or 3 m for compounds a and b, AA'BB'-type m's (2x2H) for compounds c (J: 8.5-9.0 Hz); [e] H-4'; [f] H-3',5'; [h] H-2'; [j] H-2',6'; [g,i,k] Intensity: 2H/4H/3H; [l] H-5'.

The N-acyl-β-lactam structure is proved by the two carbonyl ir bands with relatively high frequency (>1750 and >1688 cm^{-1}), the appearance of the carbon resonance line of the NHCONH group (147.0–147.5 ppm) and the ^{1}H and ^{13}C-nmr shifts characteristic of the CONH group: the shifts of the H-2,6 and substituted carbon atoms, e.g. 7.55 ppm (Table 1) and 137.2 ppm (Table 2) for 3a. (These signals can be expected at about 7.05 and 153 ppm when a (=NAr) group is present.[10])

Table 2. ^{13}C-nmr chemical shifts for compounds 3a–c, 6a–c, 7a–c and 14a–c[a,b]

Compound	C-2	C-4	C-4a	C-5	C-6,7	C-8	C-8a	C-1'	C-2',6'	C-3',5'	C-4'
3a	147.4	169.7	54.3	25.2	22.6	28.2	57.4	137.2	119.5	128.8	123.9
3b	147.5	170.1	54.7	25.5	22.9	28.4	57.9	138.2	119.8 117.7	134.9 130.1	124.2
3c	147.0	169.5	54.1	24.9	22.4	27.9	57.3	135.7	120.4	128.5[c]	128.5[c]
6a	152.5	172.1	43.9	28.0	21.6	32.9	51.3	136.3	129.3	128.3	127.4
6b	152.1	171.9	43.8	27.9	21.6	32.8	51.3	137.7	128.2 127.5	132.5 129.8[d]	129.4[d]
6c	152.2	172.0	43.8	28.0	21.6	32.8	51.2	135.2	131.1	128.3	132.1
7a	155.0	173.5	46.6	26.1	25.1 25.9	32.6	51.9	138.2	131.0.	130.1	129.1
7b	154.7	173.5	46.5	26.1	25.1 25.8	32.6	51.8	139.7	131.6[d] 129.3	134.2 131.2[d]	130.0
7c	154.6	173.3	46.5	26.0	25.0 25.8	32.5	51.7	137.0	132.8	130.0	133.7
14a	154.8	173.5	43.0	25.8	23.7 24.0	31.1	48.1	138.1	130.9	130.1	129.1
14b	154.4	173.3	42.8	25.6	23.6 23.8	31.0	48.0	139.4	131.0[d] 129.7	134.3 131.5[d]	129.9
14c	154.5	173.4	42.9	25.7	23.6 23.9	31.0	48.0	136.9	132.8	130.1	133.9

[a] In DMSO-d_6 solution; in $CDCl_3$ for compounds 3a–c; δ_{TMS}= 0 ppm. [b] At 63 MHz; at 20 MHz for compounds 7a–c and 14a–c. [c] Overlapping signals. [d] Reversed assignments may also be possible.

When heated with PPA, the N-substituted cyclopentanoazetidinones (3) and the cyclohexane derivatives (4) underwent ring transformation to yield cis- (6) and trans-condensed pyrimidinediones (7a–c), respectively.

The reaction products exhibited the following structural features proving the ring transformation. The two ν C=O ir frequencies are rather low (1720–1730 and 1670–1685 cm^{-1}), while the C-2 line is downfield shifted (152–155 ppm) for 6a–c and 7a–c. The vicinity of the electron-poor imido nitrogen instead of the donating NH group causes a characteristic paramagnetic shift of the C-2',6' and C-4' signals (by ~11 and 5 ppm, respectively) compared with those in 3a–c (Tables 1 and 2). The shift of C-1' (135.2–139.7 ppm) excludes the arylimino structure of type 5. This is also proved by the analogous ir spectra of 14a–c, 6a–c and 7a–c, as the synthesis of the former excludes the formation of structure 5 (see below).

The chemically more probable (less strained) cis-annelated structure of 6a–c follows unambiguously from the characteristic different line-widths of the

annelated hydrogens (H-4a and H-8a*): the H-4a signal splits into a quartet to a first approximation and has a signal-width of ~25 Hz ($J \cong 8$ Hz), while the halfband-width of the H-8a signal (of unresolved lines) is about 10 Hz. This also proves that H-4a is axial, while H-8a is equatorial to the alicyclic ring, suggesting the envelope conformation of the cyclopentane ring as the preferred structure, in which C-8a lies out of the plane of the other ring carbons.

This finding is in agreement with our earlier general experience that the preferred conformation of the partly or fully saturated 1,3-oxazines, thiazines and pyrimidines cis-condensed with an alicyclic ring is that in which the NH group attached to the annelated carbon is axial to the alicycle.[11-15]

The above PPA isomerization is an intramolecular transacylation, in which the attack of the more nucleophilic aryl-substituted nitrogen on the carbonyl group of the strained four-membered ring results in pyrimidinediones via ring enlargement. As expected, the ring transformation of the cyclopentane-cis-condensed compounds 3 is accompanied by retention, while that of the cyclohexane derivatives 4 proceeds with inversion. Our earlier experiments[16] showed that cyclopentane-trans-condensed six-membered 1,3-heterocycles could be prepared only in exceptional cases by the cyclization of trans-1,2-disubstituted 1,3-bifunctional cyclopentane derivatives, since the formation of a cyclopentane-trans-fused six-membered hetero ring involves considerable changes in the bond lengths and bond angles. The cis isomers of the cyclopentane-condensed saturated 1,3-heterocycles are therefore more stable than the trans isomers. In contrast, the homologous cyclohexane-trans-condensed derivatives are more stable than the cis isomers.

Isocyanate substitution of the azetidinones and subsequent ring transformation provides a new pathway for the preparation of cyclopentane-cis- (6) and cyclohexane-trans-condensed (7) dihydrouracils.

The related aromatic quinazolinediones could be prepared from anthranilic acid with urea or isocyanates.[17-19] The 3-unsubstituted analogues were made from β-ketoesters and thiourea, followed by elimination of the thioxo group.[20] The cis compound 14a (Scheme 2) and its 6-unsaturated analogue have been prepared by Kricheldorf from β-isocyanatocarboxylic acid trimethylsilyl esters.[21]

The aromatic analogues of the cyclohexane-condensed pyrimidinediones have also been prepared from imino-1,3-oxazinones of type 5 with PPA.[22] We attempted to prepare compounds 5 from cis-2-amino-1-cyclohexanecarboxylic acid (9) with isocyanates, but this failed.

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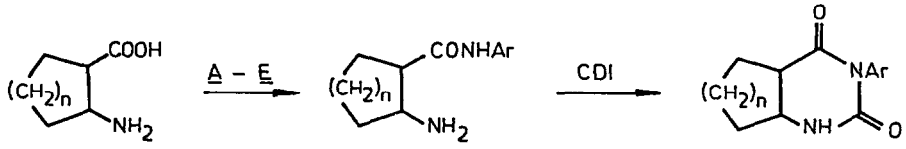
* For easier comparison of analogous spectroscopic data, the annelated carbon atom vicinal to the nitrogen is also denoted as 8a in the cyclopentane derivatives. For the same reason, the urea-carbon is denoted as C-2 (see Scheme 1).

For the proof of the presumed structures (6 and 7) and the unambiguous exclusion of structure 5, the trans derivatives 7a–c and the cis compounds 6 and 14 were also prepared from the amino acid 10[23], or from 8[24] and 9[23] through the cis (11 and 12) and trans (13) 2-amino-1-cycloalkanecarboxamides[25] by independent synthesis (Scheme 2). The amino group in 8–10 was benzyloxycarbonylated, the mixed anhydride was formed with isobutyl chloroformate, and aminolysis furnished the carboxamides. The protective group was removed with HBr–AcOH, and the 2-amino-1-carboxamides[25] (11–13) were cyclized to dihydrouracils 6, 7 and 14 with 1,1'-carbonyldiimidazole (CDI).



8: cis (n = 1) 11a–c (n = 1) 6a–c (n = 1)
9: cis (n = 2) 12a–c (n = 2) 14a–c (n = 2)
10: trans (n = 2) 13a–c (n = 2) 7a–c (n = 2)

A: $PhCH_2OCOCl$; B: ClCOOBu-i; C: $ArNH_2$; D: HBr/AcOH; E: Na_2CO_3
a: Ar = C_6H_5; b: Ar = C_6H_4Cl(m); c: Ar = C_6H_4Cl(p)

Scheme 2

Comparison of the spectral data on compounds 7a–c and 14a–c unambiguously proved the quinazolidinedione structure and the trans- (7) or cis-annelation (14). The quinazolidinedione structure is evident from the similar carbonyl frequencies (Table 1). The intervals characteristic for isomers 7 and 14 almost coincide (1720–1730 and 1670–1688 cm^{-1}, and 1726–1735 and 1686–1687 cm^{-1}). The cis-annelation of the isomers 14a–c is proved, for example, by the upfield shifts of the C-4a and C-8a lines (by about 3.5 and 4.0 ppm), which is a consequence of the steric compression shift.[26]

The practically identical halfband-width of the H-4a and H-8a ^{1}H-nmr signals for the cis isomers 14a–c, and the downfield shift of both signals compared with those for the trans pairs, show that these compounds are flexible; in solution, a roughly 1:1 equilibrium of the N-out and N-in conformers is attained: neither conformation is preferred. Katritzky et al. concluded the same equilibrium in the case of cis-5,6-tetramethylenedihydrouracil.[27] We found ratios of approximately 3:2 and 3:1 for the N-in and N-out forms, respectively, in the related 2-arylperhydroquinazolin-4-ones,[28] depending on the configuration of C-2.

The lack of a preferred conformation in 14a–c could be explained by the decreasing effect of the strained hetero ring on the inversion potential and by the carbonyl group being smaller than the methylene group. The former in-

creases the relative stability of the N-out conformer, which is unfavourable in the 4-methylene analogues (there is no steric hindrance as between H-4_{ax} and H-6_{ax} in the 4-methylene analogues). It is plausible that our general finding that the cis-annelated isomers of the 4-methylene derivatives are conformative homogeneous systems, and that the N-in or the corresponding conformation is always preferred, is not valid for the 4-carbonyl compounds reported here.

The higher ring strain decreases the stability of the cis-annelated structures compared with the trans pairs, and thus the ring enlargement can be rationalized in terms of the cis-trans isomerization.

For the aromatic derivatives, the 2-imino-1,3-oxazinone structural isomers of type 5 and the quinazolinediones exist, while among the cycloalkane-condensed analogues the dihydrouracils 6, 7 and 14 and the arylcarbamoyl-azetidinones 3 and 4 are stable. The PPA isomerization could take place via splitting of the axial C-N annelation bond of the dihydrouracil 14 formed and the more stable trans-condensed pyrimidinediones are formed by nitrogen reattack on the carbenium ion. In the cyclopentane-condensed dihydrouracils (6), no ring splitting takes place, because the cis form is the more stable.

This mechanism is supported by the transformation of 14c to the trans-condensed derivative 7c on warming with PPA, whereas the cyclopentane homologue 6 could not be isomerized to the trans compound.

In our earlier experiments on the norbornane analogues,[2] the azetidinone--pyrimidinedione ring transformation took place without inversion, similarly as for the cyclopentane derivatives 3. This could be explained by similar steric reasons. Furthermore, an exo-endo-annelated structure would also be unfavourable because of the strained skeleton.

EXPERIMENTAL

M.p.s are uncorrected. Ir spectra were run in KBr discs on a Bruker IFS-113v FT spectrometer equipped with an Aspect 2000 computer. ^{1}H- and ^{13}C-nmr spectra were recorded in $CDCl_3$ or DMSO-d_6 solution in 5 or 10 mm tubes, at room temperature, on a Bruker WM 250 (^{1}H, ^{13}C) or WP 80-SY (^{13}C) FT spectrometer controlled by an Aspect 2000 computer at 250.13 (^{1}H) and 20.14 or 62.89 (^{13}C) MHz, respectively, using the deuterium signal of the solvent as the lock and TMS as internal standard. The most important measurement parameters were as follows: sweep width 5 and 5 or 15 kHz, pulse width 1 and 3.5 or 7 μs (35° flip angle), acquisition time 1.64 or 1.02 s, number of scans 4-16 and 0.5-5 K, computer memory 16 and 16 or 32 K. Complete proton noise decoupling (~1.5 or ~3 W) for the ^{13}C spectra and Lorentzian exponential multiplication for signal--to-noise enhancement were used (line width 0.7 and 2.0 or 1.0 Hz).

Preparation of 3a-c and 4a-c

cis-2-Aza-3-oxobicyclo[3.2.0]heptane (1) (1.11 g, 0.01 mol) or cis-2-aza-3-oxobicyclo[4.2.0]octane (2) (1.25 g, 0.01 mol), isocyanate (phenyl isocyanate 1.2 g, m- or p-chlorophenyl isocyanate 1.53 g, 0.01 mol) and several drops of ethanol saturated with HCl were refluxed for 12 h. After evaporation, the residue in benzene was transferred onto a silica gel column and eluted first with benzene and then with ethyl acetate. Compounds 3 and 4 were present in the benzene eluate. After evaporation, compounds 3 were crystallized from ethanol—petroleum ether. Data on the compounds are listed in Table 3. Compounds 4a-c were transformed without purification to the dihydrouracils 7a-c.

Table 3. Physical and analytical data on compounds 3a-c, 6a-c, 7a-c and 14a-c

Compound	M.p. (°C)	Yield (%)	Formula	Required (%) C	H	N	Found (%) C	H	N
3a	68-70	72	$C_{13}H_{14}N_2O_2$	67.81	6.13	12.17	68.73	6.22	12.55
3b	57-60	63	$C_{13}H_{13}N_2ClO_2$	58.99	4.95	10.58	59.23	5.27	10.37
3c	105-107	76	$C_{13}H_{13}N_2ClO_2$	58.99	4.95	10.58	59.97	5.04	10.46
6a	255-257	52	$C_{13}H_{14}N_2O_2$	67.81	6.13	12.17	68.07	6.22	12.11
6b	269-271	45	$C_{13}H_{13}N_2ClO_2$	58.99	4.95	10.58	59.29	5.08	10.53
6c	293-295	57	$C_{13}H_{13}N_2ClO_2$	58.99	4.95	10.58	58.88	5.07	10.44
7a	261-263	42	$C_{14}H_{16}N_2O_2$	68.83	6.60	11.47	69.01	6.72	11.32
7b	249-251	40	$C_{14}H_{15}N_2ClO_2$	60.32	5.42	10.05	61.02	5.61	10.01
7c	235-237	49	$C_{14}H_{15}N_2ClO_2$	60.32	5.42	10.05	60.12	5.58	9.96
14a	145-147	54	$C_{14}H_{16}N_2O_2$	68.83	6.60	11.47	69.04	6.72	11.16
14b	124-126	51	$C_{14}H_{15}N_2ClO_2$	60.32	5.42	10.05	61.01	5.64	9.92
14c	118-120	59	$C_{14}H_{15}N_2ClO_2$	60.32	5.42	10.05	60.87	5.29	10.14

Conversion of 3a-c and 4a-c to 6a-c and 7a-c

Compounds 3a-c and 4a-c (1.0 g) were heated with PPA (30 ml) at 150 °C for 30 min. The cooled mixture was then poured into ice-water (100 ml) and the precipitate was filtered. After extraction of the aqueous phase with $CHCl_3$, the organic phase was dried (Na_2SO_4) and evaporated, and the residue was combined with the solid, transferred onto a silica gel column and eluted first with benzene then with ethyl acetate. The residue of the latter was crystallized from ethanol. Mp's of compounds 6a-c and 7a-c are listed in Table 3.

Preparation of 6a–c, 7a–c and 14a–c with carbonyldiimidazole

Carboxamides[23] (11b,c: 2.40 g, 12a and 13a: 2.53 g, 12c and 13c: 2.53 g, 0.01 mol) and 1,1'-carbonyldiimidazole (6.48 g, 0.04 mol) in benzene (30 ml) were refluxed for 8 h. The separated solid was removed by suction and washed with water. After washing and drying (Na_2SO_4), the benzene phase was evaporated off. The residue and the solid were combined and crystallized from ethanol. Yields, mp's and analytical data on compounds 6a–c, 7a–c and 14a–c are listed in Table 3.

Conversion of 14c to 7c

14c (1.0 g) in PPA (30 ml) was heated at 150 °C for 30 min. The further procedure was as above.

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