A NOVEL ONE POT SYNTHESIS OF 1,3,7-TRIARYL-1,2,3,4-TETRAHYDRO-4-OXO-5-PHENYL-2-THIOXO-5H-PYRANO[2,3-d]PYRIMIDINES

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Abstract - A novel one pot synthesis of 1,3,7-triaryl-1,2,3,4-tetrahydro-4-oxo-5-phenyl-2-thioxo-5H-pyrano[2,3-d]pyrimidines has been described. It utilizes the reaction of different thiobarbituric acids with chalcones.

Pyrans and pyrimidines individually or in combination possess significant biological properties.[1-12] A variety of routes have been described in literature for the synthesis of pyrano[2,3-d]pyrimidines.[13-26] The 2-oxo analogues of the title compounds have been prepared earlier[13] by a two step method which consists in the Michael addition of barbituric acids to chalcones and cyclisation of the formed diketo intermediate compounds.

In continuation of our work[26] on pyrano[2,3-d]pyrimidines, we herein report a new and one step synthesis of 1,3,7-triaryl-1,2,3,4-tetrahydro-4-oxo-5-phenyl-2-thioxo-5H-pyrano[2,3-d]pyrimidines in good yields (85-95%). Thus, the reaction of 1,3-(diaryl)thiobarbituric acids (1-7) with chalcone (1,3-diphenyl-2-propen-1-one) (8a) or p-methoxychalcone (8b) in acetic acid in the presence of phosphorous pentoxide at reflux temperature results in cyclocondensation to give the required 1,3,7-triaryl-1,2,3,4-tetrahydro-4-oxo-5-phenyl-2-thioxo-5H-pyrano[2,3-d]pyrimidines (16a-22a, 16b-22b). The structures were confirmed on the basis of ^{1}H-nmr spectral data which showed, besides usual signals, two doublets at around δ 4.52 (d, J=5 Hz, 1H, H-5) and around 5.99 (d, J=5 Hz, 1H, H-6). In this case, the reaction is believed to take place through the carbonium ion formation as shown below (Scheme 1). Alternatively, the title compounds have also been obtained in a two step process.[13] In this case, the reaction of thiobarbituric acids (1-7) with (8) in methanol in the presence of triethylamine at reflux temperature

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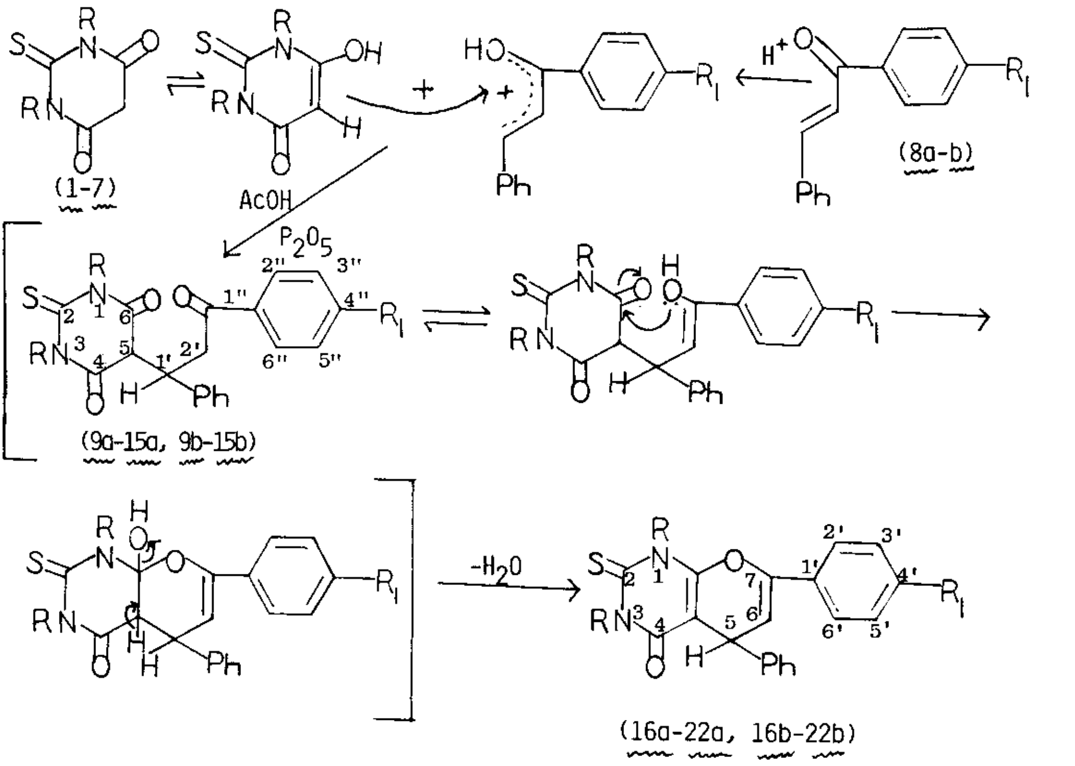
results in Michael type of addition to give 5-(2'-aroyl-1'-phenylethyl)-2-thiobarbituric acids (9a-15a, 9b-15b). These products gave positive 2,4-dinitrophenylhydrazine test and their structures were assigned on the basis of their ^{1}H-nmr spectra which showed, besides the usual signals, a multiplet at δ 4.15-4.92 integrating for three protons (CH_2-2' and H-5) and another multiplet at δ 3.90 integrating for one proton (H-1').



For,

	R		R		R_1
1, 9, 16	H	5, 13, 20	o-$CH_3C_6H_4$	8a	H
2, 10, 17	C_6H_5	6, 14, 21	m-$CH_3C_6H_4$	8b	OCH_3
3, 11, 18	o-$CH_3OC_6H_4$	7, 15, 22	p-ClC_6H_4		
4, 12, 19	m-$CH_3OC_6H_4$				

(Scheme 1)

The open chain compounds 9a-15a and 9b-15b on refluxing in glacial acetic acid in the presence of phosphorous pentoxide gave the required 16a-22a and 16b-22b. It is believed that the reaction proceeds via Michael addition of thiobarbituric acid to chalcone leading to the formation of the open chain compound which cyclises in acidic medium to give the required product.

EXPERIMENTAL

All melting points are uncorrected. ^{1}H-Nmr spectra were recorded on Perkin Elmer R-32 (90 MHz) instrument using TMS as the internal standard (chemical shifts in δ, ppm).

(i) One step method

5,7-Diphenyl-1,2,3,4-tetrahydro-4-oxo-2-thioxo-5H-pyrano[2,3-d]pyrimidine(16a) : Typical procedure

To a solution of thiobarbituric acid (1, 0.72 g, 0.005 mol) and chalcone (8a, 1.04 g, 0.005 mol) in glacial acetic acid (16 ml) was added phosphorous pentoxide (4 g) and the contents were refluxed for 20 min with continuous stirring. The reaction mixture was cooled and poured onto crushed ice. The solid thus separated was filtered, washed with dilute acetic acid (5%, 20 ml), dried and crystallised from ethanol to give 16a as white crystals (1.5 g, yield 95%), mp 290-291°C; ^{1}H-nmr (DMSO-d_6) δ : 4.52 (d, J=5 Hz, 1H, H-5), 5.99 (d, J=5 Hz, 1H, H-6), 7.32-8.10 (m, 10H, H-Ar). Anal. Calcd for $C_{19}H_{14}N_2O_2S$: C, 68.26; H, 4.19; N, 8.38. Found : C, 68.14; H, 4.17; N, 8.24.

Compounds 17a-22a and 16b-22b were obtained in a similar way.

17a : Crystallised from ethanol as white crystals (yield 80%), mp 210-211°C; ^{1}H-nmr($CDCl_3$) δ : 4.95 (d, J=5 Hz, 1H, H-5), 5.99 (d, J=5 Hz, 1H, H-6), 7.10-8.00 (m, 20H, H-Ar). Anal. Calcd for $C_{31}H_{22}N_2O_2S$: C, 76.54; H, 4.52; N, 5.76. Found : C, 76.44; H, 4.62; N, 5.80.

18a : Crystallised from ethanol as light yellow crystals (yield 85%), mp 220-221°C; ^{1}H-nmr($CDCl_3$) δ : 4.00 (s, 6H, 2xOCH_3), 4.90 (d, J=5 Hz, 1H, H-5), 6.12 (d, J=5 Hz, 1H, H-6), 7.13-7.91 (m, 18H, H-Ar). Anal. Calcd for $C_{33}H_{26}N_2O_4S$:

C, 72.52; H, 4.76; N, 5.13. Found : C, 72.29; H, 4.68; N, 5.05.

19a : Crystallised from ethanol as light yellow crystals (yield 89%), mp 180-181°C; ^{1}H-nmr($CDCl_3$) δ : 4.00 (s, 6H, 2xOCH_3), 4.95 (d, J=5 Hz, 1H, H-5), 6.00 (d, J=5 Hz, 1H, H-6), 6.95-8.15 (m, 18H, H-Ar). Anal. Calcd for $C_{33}H_{26}N_2O_4S$: C, 72.52; H, 4.76; N, 5.13. Found : C, 72.33; H, 4.67; N, 5.21.

20a : Crystallised from ethanol as light yellow crystals (yield 86%), mp 198-199°C; ^{1}H-nmr($CDCl_3$) δ : 2.33 (s, 6H, 2xCH_3), 4.95 (d, J=5 Hz, 1H, H-5), 6.12 (d, J=5 Hz, 1H, H-6), 7.30-8.15 (m, 18H, H-Ar). Anal. Calcd for $C_{33}H_{26}N_2O_2S$: C, 77.04; H, 5.05; N, 5.44. Found : C, 77.10; H, 5.01; N, 5.36.

21a : Crystallised from ethanol as pale yellow crystals (yield 88%), mp 200-201°C; ^{1}H-nmr($CDCl_3$) δ : 2.35 (s, 6H, 2xCH_3), 4.75 (d, J=5 Hz, 1H, H-5), 5.82 (d, J=5 Hz, 1H, H-6), 6.81-7.82 (m, 18H, H-Ar). Anal. Calcd for $C_{33}H_{26}N_2O_2S$: C, 77.04; H, 5.05; N, 5.44. Found : C, 77.20; H, 5.02; N, 5.41.

22a : Crystallised from ethanol as light yellow crystals (yield 89%), mp 180-181°C; ^{1}H-nmr($CDCl_3$) δ : 4.75 (d, J=5 Hz, 1H, H-5), 5.85 (d, J=5 Hz, 1H, H-6), 6.99-7.91 (m, 18H, H-Ar). Anal. Calcd for $C_{31}H_{20}N_2O_2SCl_2$: C, 67.02; H, 3.60; N, 5.04. Found : C, 67.14; H. 3.56; N, 5.12.

16b : Crystallised from methanol as pink crystals (yield 92%), mp 244-245°C; ^{1}H-nmr($CDCl_3$) δ : 4.10 (s, 3H, OCH_3), 4.82 (d, J=5 Hz, 1H, H-5), 5.93 (d, J=5 Hz, 1H, H-6), 7.24 (d, J=9.5 Hz, 2H, H-3' and H-5'), 7.33-8.12 (m, 5H, H-Ar), 8.40 (d, J=9.5 Hz, 2H, H-2' and H-6'). Anal. Calcd for $C_{20}H_{16}N_2O_3S$: C, 65.93; H, 4.41; N, 7.69. Found : C, 65.74; H, 4.32; N, 7.54.

17b : Crystallised from methanol as white crystals (yield 95%), mp 230-232°C; ^{1}H-nmr($CDCl_3$) δ : 3.95 (s, 3H, OCH_3), 4.92 (d, J=5 Hz, 1H, H-5), 5.93 (d, J=5 Hz, 1H, H-6), 6.91 (d, J=9.5 Hz, 2H, H-3' and H-5'), 7.14-8.05 (m, 17H, H-Ar). Anal. Calcd for $C_{32}H_{24}N_2O_3S$: C, 74.42; H, 4.65; N, 5.42. Found : C, 74.33; H, 4.81; N, 5.34.

18b : Crystallised from methanol as light yellow crystals (yield 92%), mp 184-185°C; ^{1}H-nmr($CDCl_3$) δ : 3.87, 3.92 & 3.98 (3xs, 9H, 3xOCH_3), 4.84 (d, J=5 Hz, 1H, H-5), 5.82 (d, J=5 Hz, 1H, H-6), 6.93 (d, J=9.5 Hz, 2H, H-3' and H-5'), 7.05-7.92 (m, 15H, H-Ar). Anal. Calcd for $C_{34}H_{28}N_2O_5S$: C, 70.83; H, 4.86; N, 4.86. Found : C, 70.64; H, 4.72; N, 4.66.

19b : Crystallised from methanol as light yellow crystals (yield 95%), mp 165-166°C; ^{1}H-nmr($CDCl_3$) δ : 3.86, 3.91 & 3.96 (3xs, 9H, $3xOCH_3$), 4.93 (d, J=5 Hz, 1H, H-5), 5.92 (d, J=5 Hz, 1H, H-6), 6.83-8.32 (m, 17H, H-Ar). Anal. Calcd for $C_{34}H_{28}N_2O_5S$: C, 70.83; H, 4.86; N, 4.86. Found : C, 70.71; H. 4.80; N, 4.71.

20b : Crystallised from methanol as cream crystals (yield 85%), mp 209-210°C; ^{1}H-nmr($CDCl_3$) δ : 2.38 & 2.45 (2xs, 6H, $2xCH_3$), 3.94 (s, 3H, OCH_3), 4.91 (d, J=5 Hz, 1H, H-5), 5.92 (d, J=5 Hz, 1H, H-6), 6.93 (d, J=9.5 Hz, 2H, H-3' and H-5'), 7.15-7.95 (m, 15H, H-Ar). Anal. Calcd for $C_{34}H_{28}N_2O_3S$: C, 75.00; H, 5.15; N, 5.15. Found : C, 74.82; H, 5.05; N, 5.12.

21b : Crystallised from methanol as yellow crystals (yield 87%), mp 129-130°C; ^{1}H-nmr($CDCl_3$) δ : 2.54 & 2.59 (2xs, 6H, $2xCH_3$), 3.92 (s, 3H, OCH_3), 4.93 (d, J=5 Hz, 1H, H-5), 5.90 (d, J=5 Hz, 1H, H-6), 6.91 (d, J=9.5 Hz, 2H, H-3' and H-5'), 7.12-8.41 (m, 15H, H-Ar). Anal. Calcd for $C_{34}H_{28}N_2O_3S$: C, 75.00; H, 5.15; N, 5.15. Found : C, 74.91; H, 5.01; N, 5.21.

22b : Crystallised from methanol as light yellow crystals (yield 85%), mp 256-257°C; ^{1}H-nmr($CDCl_3$) δ : 3.90 (s, 3H, OCH_3), 4.82 (d, J=5 Hz, 1H, H-5), 5.81 (d, J=5 Hz, 1H, H-6), 6.81-8.34 (m, 17H, H-Ar). Anal. Calcd for $C_{32}H_{22}N_2O_3SCl_2$: C, 65.64; H, 3.76; N, 4.78. Found : C, 65.54; H, 3.82; N, 4.63.

(ii) Two step method

5-(2'-Benzoyl-1'-phenylethyl)-2-thiobarbituric acid (9a) : Typical procedure

Thiobarbituric acid (1, 0.72 g, 0.005 mol), chalcone (8a, 1.04 g, 0.005 mol) and dry triethylamine (15 ml) in methanol (50 ml) were refluxed for 5-6 h. The reaction mixture was then cooled, poured onto crushed ice and neutralised with dilute hydrochloric acid. The product thus separated was filtered, dried and crystallised from benzene as light yellow crystals of 9a (1.1 g, yield 68%), mp 300-301°C; ^{1}H-nmr (DMSO-d_6) δ : 3.90 (m, 1H, H-1'), 4.15-4.90 (m, 3H, CH_2-2' and H-5), 7.31-7.93 (m, 8H, H-Ar), 8.25 (m, 2H, H-2" and H-6"). Anal. Calcd for $C_{19}H_{16}N_2O_3S$: C, 64.77; H, 4.54; N, 7.95. Found : C, 64.67; H, 4.56; N, 7.90. Compounds 10a-15a and 9b-15b were synthesised in a similar way.

10a : Crystallised from benzene as yellow crystals (yield 65%), mp 230-231°C; ^{1}H-nmr($CDCl_3$) δ : 3.85 (m, 1H, H-1'), 4.51-4.92 (m, 3H, CH_2-2' and H-5), 6.85-8.05 (m, 18H, H-Ar), 8.25 (m, 2H, H-2" and H-6"). Anal. Calcd for $C_{31}H_{24}N_2O_3S$: C, 73.81; H, 4.76; N, 5.55. Found : C, 73.72; H, 4.80; N, 5.49.

11a : Crystallised from benzene as yellow crystals (yield 60%), mp 190-191°C; ^{1}H-nmr($CDCl_3$) δ : 3.50 (m, 1H, H-1'), 3.75 (s, 6H, 2xOCH_3), 4.10-4.71 (m, 3H, CH_2-2' and H-5), 6.62-7.81 (m, 16H, H-Ar), 8.15 (m, 2H, H-2" and H-6"). Anal. Calcd for $C_{33}H_{28}N_2O_5S$: C, 70.21; H, 4.96; N, 4.96. Found : C, 70.05; H, 4.89; N, 4.93.

12a : Crystallised from benzene as yellow crystals (yield 65%), mp 158-159°C; ^{1}H-nmr($CDCl_3$) δ : 3.45 (m, 1H, H-1'), 3.99 (s, 6H, 2xOCH_3), 4.10-4.52 (m, 3H, CH_2-2' and H-5), 6.70-7.64 (m, 16H, H-Ar), 8.05 (m, 2H, H-2" and H-6"). Anal. Calcd for $C_{33}H_{28}N_2O_5S$: C, 70.21; H, 4.96; N, 4.96. Found : C, 70.14; H, 4.88; N, 4.92.

13a : Crystallised from benzene as yellow crystals (yield 68%), mp 190-191°C; ^{1}H-nmr($CDCl_3$) δ : 2.15 (s, 6H, 2xCH_3), 3.75 (m, 1H, H-1'), 4.50-4.91 (m, 3H, CH_2-2' and H-5), 7.20-7.95 (m, 16H, H-Ar), 8.30 (m, 2H, H-2" and H-6"). Anal. Calcd for $C_{33}H_{28}N_2O_3S$: C, 74.43; H, 5.26; N, 5.26. Found : C, 74.32; H, 5.32; N, 5.09.

14a : Crystallised from benzene as yellow crystals (yield 67%), mp 160-161°C; ^{1}H-nmr($CDCl_3$) δ : 2.32 (s, 6H, 2xCH_3), 3.63 (m, 1H, H-1'), 4.05-4.54 (m, 3H, CH_2-2' and H-5), 7.20-7.93 (m, 16H, H-Ar), 8.31 (m, 2H, H-2" and H-6"). Anal. Calcd for $C_{33}H_{28}N_2O_3S$: C, 74.43; H, 5.26; N, 5.26. Found : 74.29; H, 5.19; N, 5.18.

15a : Crystallised from benzene as yellow crystals (yield 67%), mp 240-241°C; ^{1}H-nmr($CDCl_3$) δ : 3.70 (m, 1H, H-1'), 4.20-4.72 (m, 3H, CH_2-2' and H-5), 6.60-

7.71 (m, 16H, H-Ar), 8.30 (m, 2H, H-2" and H-6"). Anal. Calcd for $C_{31}H_{22}N_2O_3SCl_2$: C, 64.92; H, 3.84; N, 4.88. Found : C, 64.81; H, 3.78; N, 4.69.

9b : Crystallised from benzene as white crystals (yield 70%), mp 221-222°C; ^{1}H-nmr($CDCl_3$) δ : 3.70 (m, 1H, H-1'), 4.15-4.82 (m, 3H, CH_2-2' and H-5), 3.95 (s, 3H, OCH_3), 7.12 (d, J=9.5 Hz, 2H, H-3" and H-5"), 7.40 (s, 5H, C_6H_5), 8.15 (d, J=9.5 Hz, 2H, H-2" and H-6"). Anal. Calcd for $C_{20}H_{18}N_2O_4S$: C, 62.82; H, 4.71; N, 7.33. Found : C, 62.69; H, 4.80; N, 7.26.

10b : Crystallised from benzene as pale yellow crystals (yield 71%), mp 215-216°C; ^{1}H-nmr($CDCl_3$) δ : 3.80 (m, 1H, H-1'), 4.00 (s, 3H, OCH_3), 4.31-4.82 (m, 3H, CH_2-2' and H-5), 7.30 (d, J=9.5 Hz, 2H, H-3" and H-5"), 7.50 and 7.80 (each m, 15H, 3xC_6H_5), 8.40 (d, J=9.5 Hz, 2H, H-2" and H-6"). Anal. Calcd for $C_{32}H_{26}N_2O_4S$: C, 71.91; H, 4.87; N, 5.24. Found : C, 71.87; H, 4.88; N, 5.27.

11b : Crystallised from benzene as light yellow crystals (yield 65%), mp 192-193°C; ^{1}H-nmr($CDCl_3$) δ : 3.62 (m, 1H, H-1'), 3.88, 3.93 & 3.98 (3xs, 9H, 3xOCH_3), 4.05-4.65 (m, 3H, CH_2-2' and H-5), 6.91-7.72 (m, 15H, H-3", H-5", C_6H_5 and 2xN-Ar), 8.15 (d, J=9.5 Hz, H-2" and H-6"). Anal. Calcd for $C_{34}H_{30}N_2O_6S$: C, 68.68; H, 5.05; N, 4.71. Found : C, 68.59; H, 4.96; N, 4.68.

12b : Crystallised from benzene as cream crystals (yield 70%), mp 60-61°C; ^{1}H-nmr ($CDCl_3$) δ : 3.52 (m, 1H, H-1'), 3.86, 3.91 & 3.96 (3xs, 9H, 3xOCH_3), 4.02-4.62 (m, 3H, CH_2-2' and H-5), 6.90-7.92 (m, 15H, H-3", H-5", C_6H_5 and 2xN-Ar), 8.30 (d, J=9.5 Hz, 2H, H-2" and H-6"). Anal. Calcd for $C_{34}H_{30}N_2O_6S$: C, 68.68; H, 5.05; N, 4.71. Found : C, 68.58; H, 5.06; N, 4.67.

13b : Crystallised from benzene as cream crystals (yield 72%), mp 79-80°C; ^{1}H-nmr($CDCl_3$) δ : 2.20 & 2.25 (2xs, 6H, 2xCH_3), 3.80 (m, 1H, H-1'), 4.00 (s, 3H, OCH_3), 4.21-4.43 (m, 3H, CH_2-2' and H-5), 7.23 (d, J=9.5 Hz, 2H, H-3" and H-5"), 7.31-8.52 (m, 15H, H-2", H-6", C_6H_5 and 2xN-Ar). Anal. Calcd for $C_{34}H_{30}N_2O_4S$: C, 72.59; H, 5.34; N, 4.98. Found : C, 72.55; H, 5.42; N, 4.90.

14b : Crystallised from benzene as light yellow crystals (yield 70%). mp 111-112°C; ^{1}H-nmr($CDCl_3$) δ : 2.38 & 2.43 (2xs, 6H, 2xCH_3), 3.62 (m, 1H, H-1'), 3.95 (s, 3H,

OCH_3), 4.05-4.61 (m, 3H, CH_2-2' and H-5), 6.91-7.82 (m, 15H, H-3", H-5", C_6H_5 and 2xN-Ar), 8.25 (d, J=9.5 Hz, 2H, H-2" and H-6"). Anal. Calcd for $C_{34}H_{30}N_2O_4S$: C, 72.59; H, 5.34; N, 4.98. Found : C, 72.54; H, 5.38; N, 4.91.

15b : Crystallised from benzene as light yellow crystals (yield 73%), mp 81-83°C; ^{1}H-nmr ($CDCl_3$) δ : 3.70 (m, 1H, H-1'), 3.92 (s, 3H, OCH_3), 4.10-4.61 (m, 3H, CH_2-2' and H-5), 6.90-8.32 (m, 17H, H-Ar). Anal. Calcd for $C_{32}H_{24}N_2O_4SCl_2$: C, 63.68; H, 3.98; N, 4.64. Found : C, 63.59; H, 4.04; N, 4.57.

5,7-Diphenyl-1,2,3,4-tetrahydro-4-oxo-2-thioxo-5H-pyrano[2,3-d]pyrimidine (16a) : Typical procedure

To a solution of phosphorous pentoxide (4 g) in glacial acetic acid (16 ml) was added the adduct 9a (1 g) and the mixture was refluxed for 1 h. The reaction mixture was cooled and poured onto crushed ice. The product separated was filtered, washed with dil. acetic acid (5%, 20 ml), dried and crystallised from ethanol as white crystals (0.95 g, yield 95%), mp 290-291°C. Similar cyclization of 10a-15a and 9b-15b gave 17a-22a and 16b-22b respectively in 85-95% yield.

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