Pergamon

Tetrahedron Vol. 51, No. 34, pp. 9385-9392, 1995
Elsevier Science Ltd
Printed in Great Britain
0040-4020/95 $9.50+0.00

0040-4020(95)00545-5

Cycloaddition Reactions of 5H,7H-Thiazolo[3,4-c] Oxazolium-1-Oxides with Imines

Piero Dalla Croce[a], Raffaella Ferraccioli[a], and Concetta La Rosa*[,b]

[a] Dipartimento di Chimica Organica e Industriale and Centro C.N.R., V. Venezian 21, 20133 Milano - Italy

[b] Istituto di Chimica Organica, Facoltà di Farmacia, V. Venezian 21, 20133 Milano - Italy

Abstract: The behaviour of mesoionic 5H,7H-thiazolo [3,4-c] oxazol-1-ones (**α**) towards imines (**2**) and (**3**) was studied. The cycloaddition reaction affords 7-thia-2,5-diazaspiro [3,4] octan-1-one (**4-7**) and 1H,3H-imidazo [1,5-c] thiazole (**8**) derivatives. The possible mechanism involved in the formation of products as well as the unusual rearrangement showed by spirocyclic β-lactams (**6,7**) are discussed.

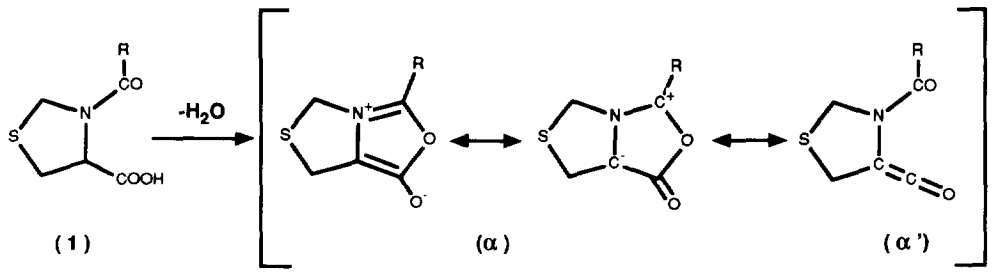
INTRODUCTION

In some previous communications we reported the synthesis of various heterocyclic systems through the 1,3-dipolar cycloaddition reactions of 1,3-oxazolium-5-oxides, commonly known as Münchnones, with different dipolarophiles.[1-5] In these studies we used monocyclic Münchnones deriving from linear N-acyl-α-amino acids.

In the present work we extended our studies to the 5H,7H-thiazolo [3,4-c] oxazolium-1-oxides (α), bicyclic mesoionic compounds deriving from N-acyl-(R)-thiazolidine-4-carboxylic acids (**1**) (Scheme 1). These bicyclic mesoionic compounds have been scarcely employed as 1,3-dipoles in cycloaddition reactions: the only examples reported deal with their addition to dimethyl acetylenedicarboxylate[6-9] and to α-chloroacrylonitrile[10] affording 1H,3H-pyrrolo [1,2-c] thiazole derivatives.

In connection with our previous works concerning the use of imines as dipolarophiles[4,5,11] we examined the reactivity of N-(phenylmethylene)aniline (**2**) and N-(phenylmethylene)benzenesulfonamide (**3**) towards (α).The effort of our work was to verify the capability of mesoionic 5H,7H-thiazolo [3,4-c] oxazol-1-ones to react with a C=N double bond giving, through a 1,3-dipolar cycloaddition , the 1H,3H-imidazo[1,5-c] thiazole system.

SCHEME 1



RESULTS AND DISCUSSION

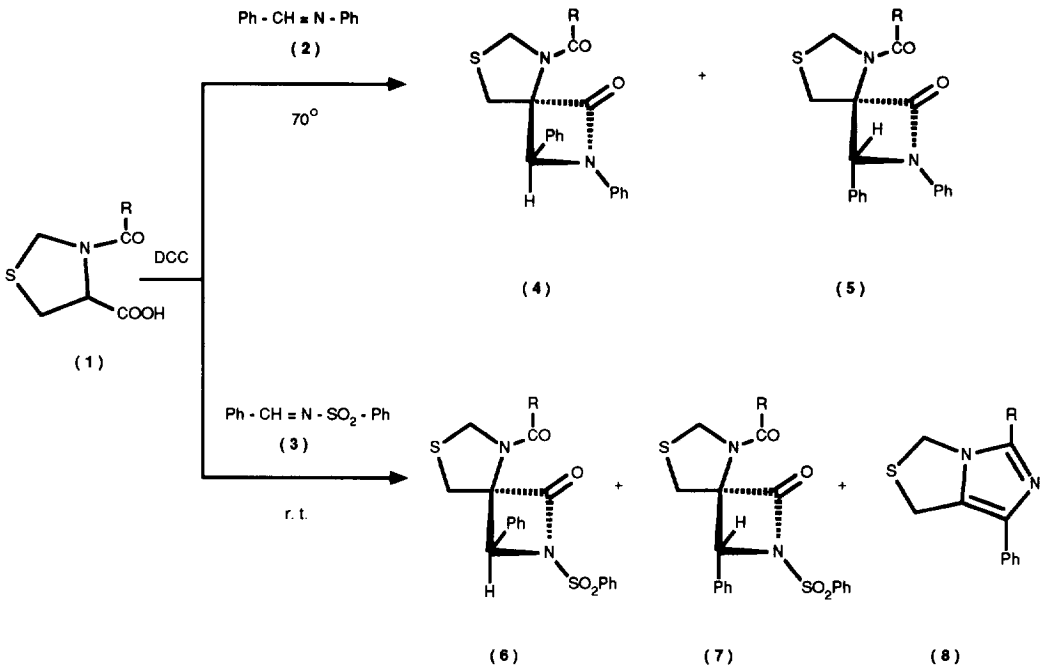
Owing to their high reactivity the mesoionic compounds (α) were prepared in situ starting from the corresponding N-acyl-(R)-thiazolidine-4-carboxylic acids (**1a-c**) and N,N'-dicyclohexylcarbodiimide[12a] as dehydrating agent. The reactions with imines (**2**) and (**3**) were performed in tetrahydrofuran solution at room temperature or at 70° C according to the type of imines. Filtration of dicyclohexylurea and evaporation of the solvent gave a mixture of products which were separated by column chromatography and further purified by crystallization. Analytical, physical and spectroscopic data of the isolated products are summarized in Tables 1 and 2.

As shown in Scheme 2 the reaction of (**1a,b**) and imine (**2**) led to a mixture of diastereoisomeric 7-thia-2,5-diazaspiro [3,4] octan-1-one (**4**) and/or (**5**) while the reaction of (**1a-c**) with imine (**3**) afforded beside spirocyclic β-lactams (**6**) and (**7**) also a variable amount of the 1H,3H-imidazo [1,5-c] thiazoles (**8**)[12b].

Structures (**4**)/(**5**) and (**6**)/(**7**) were assigned on the basis of their analytical and spectroscopic data : they are two couples of diastereoisomers which differ only in the configuration at spiranic atom. The I.R. spectra of each adduct showed two bands at 1750-1805 and 1635-1670 cm^{-1} which agree with a β-lactamic and an amidic carbonyl group respectively. The 300 MHz ^{13}C N.M.R. spectra of each product showed the signals relative to two carbonylic carbons and one sp^3 spiranic carbon. The 300 MHz ^1H N.M.R. spectra also confirmed these structures, moreover they allowed to assign relative steric configuration to the two diastereoisomers. In fact spectra of products (**5**) and (**7**) showed for the benzylic proton a signal with a chemical shift 0.3-0.5 ppm lower than that of the corresponding diastereoisomers (**4**) and (**6**) due to the deshielding effect of the N-acyl carbonyl group on this proton.

The diastereoisomeric ratio of structures (**4**)/(**5**) and (**6**)/(**7**) was determined on the basis of the ^1H-NMR analysis of the crude products mixture evaluating the integration of the benzylic proton at 5.1-5.6 δ. The ratio experimentally observed can be easily explained observing in the corresponding molecular models of (**4**) and (**6**) the remarkable steric encumbrance between the lactamic ring C-phenyl group and the thiazolidine N-acyl group.

SCHEME 2



(1, 4, 5 a)	R = CH_3	(4 a) : (5 a) = 40 : 60
(1, 5 b)	R = C_6H_5	(4 b) : (5 b) = 0 : 100
(1, 6, 7 a)	R = CH_3	(6 a) : (7 a) : (8 a) = 12 : 88 : 0
(1, 6, 7, 8 b)	R = C_6H_5	(6 b) : (7 b) : (8 b) = 1 : 29 : 70
(1, 6, 7, 8 c)	R = 4-CH_3O-C_6H_4	(6 c) : (7 c) : (8 c) = 2 : 40 : 58

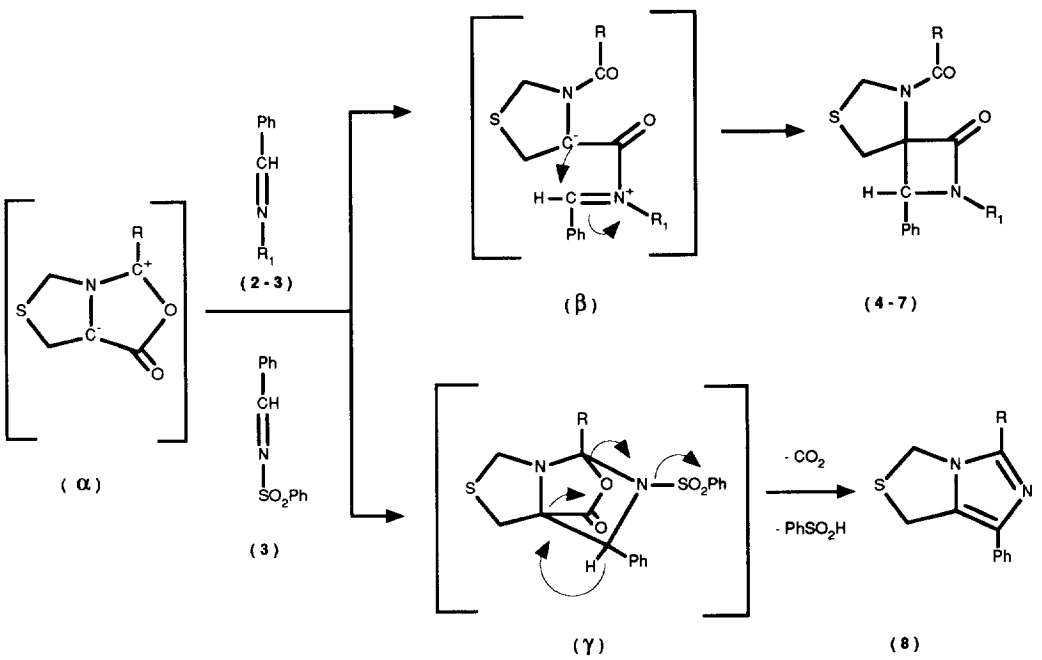
Also structures (**8**), till now unknown, were assigned on the basis of their analytical and spectroscopic data on the analogy of the 1H,3H-pyrrolo [1,2-c] thiazole system.[9] The absence of any carbonyl group in I.R. and ^{13}C N.M.R. spectra and the lack of N-phenylsulfonyl substituent agree with the assigned structure of 1H,3H-imidazo [1,5-c] thiazole .

As indicated in Scheme 2 the reaction of (**1**) with imines (**2**) and (**3**) leads to a mixture of diastereoisomeric spirocyclic β-lactams (**4-7**) and only in the case of the reaction of (**1b,c**) with imine (**3**) a third product (**8**) was isolated. On the basis of our results it comes out that the reaction is influenced by the nature of substituents on imine (phenyl or phenylsulfonyl) and on mesoionic compounds (R = methyl or aryl). Generally imine (**2**) requires higher temperatures and gives lower yields in comparison with imine (**3**) which reacts at room temperature and gives better yields. Moreover in the case of imine (**3**) the reaction with (**1b,c**) leads mainly to imidazothiazoline adducts (**8**).

To explain these results we hypothesize that the reaction occurs according to two mechanisms (Scheme 3). The diastereoisomeric spirocyclic β-lactams (**4-7**) derive from a two steps addition involving the

nucleophilic attack of nitrogen imine to the carbonyl group of mesoionic compound (α) with formation of a zwitterionic intermediate (β) followed by ring closure to (**4-7**)[5]. On the contrary the imidazothiazole derivatives (**8**) derive from a typical 1,3-dipolar cycloaddition of mesoionic (α) to the C=N double bond of imine (**3**) with formation of the tricyclic adduct (γ) from wich loss of carbon dioxide and benzenesulfinic acid gives rise to products (**8**).

SCHEME 3



Another possible mechanism for the β-lactams formation could be a [2+2] cycloaddition between imines and the ketene valence tautomer (α')[13]. The real difficulty of the N-acyl-(R)-thiazolidine-4-carboxylic acids (**1**) to undergo cyclization[14] could shift the equilibrium between the bicyclic system (α) and the corresponding ketene (α') towards this latter; therefore it is not possible to exclude the presence of the ketene tautomer in the dehydration process of N-acyl-(R)-thiazolidine-4-carboxylic acid.

The reaction paths followed by the imines (**2**) and (**3**) could depend on their different nucleophilicity and also on electronic factors, due to the different type of substituent R, that could influence the stability of the reactive intermediates.

Another difference that points out the role played by the N-phenylsulfonyl group deals with the thermal stability of (**4,5**) and (**6,7**). Compounds (**4,5**) are stable up to 200° C, whereas (**6,7**) on heating at 185-200° C give the corresponding products (**8**). This unexpected transformation, which allows the 1H,3H-imidazo[1,5-c] thiazole derivatives to be obtained starting from the N-phenylsulfonyl spirocyclic β-lactams, prompts us to investigate the possible pathway.

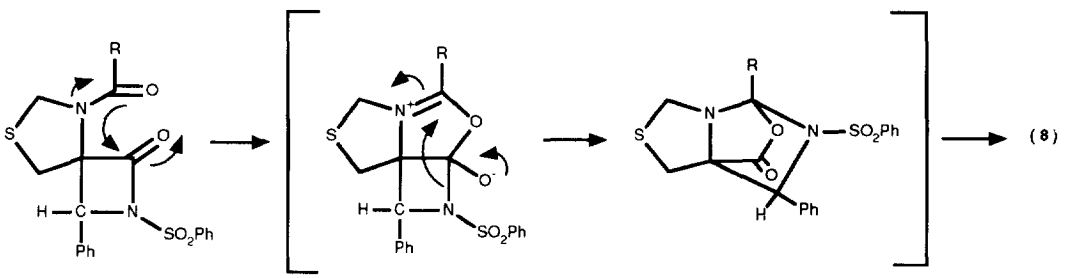
The hypothesis of a cycloreversion of (**6,7**) to starting materials followed by a [3+2] cycloaddition

reaction was ruled out since the heating of **(6b,7b)** in tetraline solution at T=190° C in the presence of N-[(4-methoxy)phenylmethylene]benzenesulfonamide did not afford a mixture of products containing the 4-methoxyphenyl group, but only **(8b)** in agreement with an intramolecular process.

Also the pathway involving the elimination of benzenesulfinic acid from the β-lactamic ring followed by ring opening to benzonitrile and (α) was excluded on the basis of the absence of mixed products in the rearrangement reaction of **(6b,7b)** performed in the presence of 4-methoxy-benzonitrile.

Therefore a possible explanation could be an intramolecular attack of the N-acyl oxygen on the lactamic carbonyl[15] followed by a rearrangement leading to the tricyclic intermediate (γ) from which products **(8)** derive (Scheme 4).

SCHEME 4



(6,7) (γ)

The different thermal stability observed between **(4/5)** and **(6/7)** could be explained taking into account the possibility of intermediate (γ) to lose beside carbon dioxide also benzenesulfinic acid leading to an aromatic product **(8)**.

The cycloaddition reaction of mesoionic 5H,7H-thiazolo [3,4-c] oxazolium-1-oxides with imines described in this work allowed to obtain new spirocyclic β-lactams N-phenyl **(4,5)** or N-phenylsulfonyl **(6,7)** derivatives and 1H,3H-imidazo [1,5-c] thiazole **(8)**, these latter either from the cycloaddition reaction or from thermal rearrangement of the corresponding spirocyclic β-lactams.

Acknowledgements. This work was supported by the Italian National Research Council (C.N.R.) Progetto Finalizzato Chimica Fine II and by MURST.

EXPERIMENTAL

General Methods. M.p.s.were measured with a Büchi apparatus and are uncorrected. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker AC 300 spectrometer. Infrared spectroscopy was performed on a Perkin-Elmer 298 spectrophotometer. MS spectra were determined on a VG Analytical 7070 EQ mass spectrometer with a VG Analytical 11/250 data system attached.

N-Acetyl-(R)-thiazolidine-4-carboxylic acid (1a): Compound **(1a)** was prepared by the method of Ratner[16] to give 84% of a white solid, m.p. 145° C (lit. 144-5° C); $[\alpha]_D^{20}$=-124.6° (c=1, MeOH); ^{1}H-NMR ($CDCl_3$) (two conformers) : δ 2.1,2.2 (3H,s,$COCH_3$), 3.2,3.5 (2H,m,S-CH_2-C), 4.5,4.8 (2H,dd,S-CH_2-N), 4.9 (1H,broad,COOH), 5.1 (1h,m,CH).

N-Benzoyl-(R)-thiazolidine-4-carboxylic acid (1b): Compound **(1b)** was prepared by the method of Neher[17] to give 95% of an amorphous colorless solid, $[\alpha]_D^{20}$=-207.2° (c=1, MeOH); ^{1}H-nmr ($CDCl_3$): δ 3.4 (2H,broad,S-CH_2-C), 4.6 (2H,broad,S-CH_2-N), 5.3 (1H,broad,CH), 6.3 (1H,broad,COOH), 7.4-7.7 (5H,m,aromatics).

N-(4-Methoxybenzoyl)-(R)-thiazolidine-4-carboxylic acid (1c): Compound **(1c)** was prepared by the same method utilized for **(1b)** giving, after purification by column chromatography (silica gel, toluene/dioxane/acetic acid = 45/10/2), 75% of an amorphous colorless solid, ^{1}H-NMR ($CDCl_3$): δ 3.3 (2H,broad,S-CH_2-C), 3.8 (3h,s,OCH_3), 4.6 (2H,broad,S-CH_2-N), 5.3 (1H,m,CH), 6.9 (2H,d,aromatics), 7.5 (2H,d,aromatics), 10.5 (1H,broad, COOH).

N-Phenylmethylene-aniline (2): Compound **(2)** was prepared according to the method of Bigelow[18] to give 83% of a white solid, m.p. 54° C (lit. 52° C).

N-Phenylmethylene-benzenesulfonamide (3): Compound **(3)** was prepared according to the method of Jennings[19] to give 69% of a white solid, m.p. 85° C (lit.[20] 80° C).

General Procedure for Cycloaddition reactions of (1a-c) with (2)/(3). To a stirred solution of **(1a-c)** (4 mmol) in tetrahydrofuran (10 ml) was added, dropwise, a solution of dicyclohexylcarbodiimide (4.4 mmol) in tetrahydrofuran (10 ml) under nitrogen. After about 15 minutes a solution of imine **(2)** or **(3)** (4 mmol) in tetrahydrofuran (5 ml) was added dropwise. The mixture was stirred at room or reflux temperature for 24 h. The N,N'-dicyclohexylurea was filtered off and after evaporation of the solvent the crude mixture was separated by column chromatography (silica gel, toluene/ethyl acetate : 90/10). Products **(4-8)** were recrystallized and identified by analytical and spectroscopic data (Tables 1 and 2).

Thermal Rearrangement of products (6)/(7). 100 mg of **(6a-c)/(7a-c)** were heated at T=185-200° C for 1h under nitrogen. The brown residue was purified by column chromatography (silica gel, toluene/ethyl acetate:90/10) affording products **(8a-c)**.

5-Methyl-7-phenyl-1H,3H-imidazo [1,5-c] thiazole (8a): yield 8%; m.p. 120° C (dec.) (n-exane/toluene); ^{1}H-NMR ($CDCl_3$): δ 2.38 (3H,s,CH_3), 4.25 (2H,s,S-CH_2-C), 4.90 (2H,s,S-CH_2-N), 7.1-7.65 (5H,m,aromatics).

5,7-Diphenyl-1H,3H-imidazo [1,5-c] thiazole (8b): yield 53%; m.p. and ^{1}H-NMR ($CDCl_3$) see Tables 1 and 2.

5-(4-Methoxyphenyl)-7-phenyl-1H,3H-imidazo [1,5-c] thiazole (8c): yield 50%; m.p. and ^{1}H-NMR ($CDCl_3$) see Tables 1 and 2.

Table 1. Yields, Physical and Analytical Data of Products (4) - (8)

N°	Yield (%)[a]	m.p. (°C) (cryst. solv.)	Molecular Formula	Elemental Analysis (%) C H N Found (Required)
4 a	7	191-193 (n-BuOH)	$C_{19}H_{18}N_2O_2S$	67.58 5.55 8.57 (67.45 5.32 8.28)
5 a	8	180-182 (n-BuOH)	$C_{19}H_{18}N_2O_2S$	67.50 5.40 8.50 (67.45 5.32 8.28)
5 b	7	170-173 (Diisopr.eth.)	$C_{24}H_{20}N_2O_2S$	71.84 4.80 7.20 (72.00 5.00 7.00)
6 a	2	193-194 d. (EtOH)	$C_{19}H_{18}N_2O_4S_2$	56.98 4.29 7.02 (56.72 4.48 6.96)
7 a	19	180-181 d. (EtOH)	$C_{19}H_{18}N_2O_4S_2$	56.41 4.66 6.67 (56.72 4.48 6.96)
6 b	1	208-210 d. (Tol.)	$C_{24}H_{20}N_2O_4S_2$	61.77 4.51 5.92 (62.07 4.31 6.03)
7 b	26	195-197 d. (Tol.)	$C_{24}H_{20}N_2O_4S_2$	61.87 4.45 5.98 (62.07 4.31 6.03)
8 b	52	211-212 d. (Tol.)	$C_{17}H_{14}N_2S$	73.00 5.05 9.84 (73.38 5.04 10.07)
6 c	1	161-163 d. (Diisopr.eth.)	$C_{25}H_{22}N_2O_5S_2$	60.52 4.20 5.58 (60.73 4.45 5.67)
7 c	19	167-168 d. (EtOH)	$C_{25}H_{22}N_2O_5S_2$	60.61 4.32 5.47 (60.73 4.45 5.67)
8 c	30	146-147 (Diisopr.eth.)	$C_{18}H_{16}N_2OS$	69.93 5.05 9.01 (70.13 5.19 9.10)

[a]Yield of pure isolated products.

Table 2. Spectroscopic Data of Products (4) - (8)

N°	^{1}H-NMR ($CDCl_3$) δ from TMS	^{13}C-NMR ($CDCl_3$) δ from TMS	I R (cm^{-1})	MS (m/e)
4 a	1.7 (s,3H,CH_3); 2.7-3.2 (dd,2H,S-CH_2-C); 4.5 (dd,2H,S-CH_2-N); 5.0 (s,1H,CH); 7.0-7.4 (m,10H,aromatics).	22.3 (CH_3); 37.6 (S-CH_2-C); 49.8 (S-CH_2-N); 70.0 (CH); 79.3 (C sp.); 162.7,168.8 (2 CO).	1670 1750	338 295
5 a	2.2 (s,3H,CH_3); 3.3-3.6 (dd,2H,S-CH_2-C); 4.1-4.5 (dd,2H,S-CH_2-N); 5.5 (s,1H,CH); 7.0-7.4 (m,10H,aromatics).	23.4 (CH_3); 32.9 (S-CH_2-C); 50.7 (S-CH_2-N); 65.4 (CH); 79.3 (C sp.); 160.0,167.7 (2 CO).	1660 1760	338 295
5 b	2.8-3.3 (dd,2H,S-CH_2-C); 4.6 (dd, 2H, S-CH_2-N); 5.8 (s,1H,CH); 7.3-7.7 (m,15H, aromatics).	----------	1660 1765	400 295
6 a	1.7 (s,3H,CH_3); 3.0-3.4 (dd,2H,S-CH_2-C); 4.0-4.5 (dd,2H,S-CH_2-N); 5.1 (s,1H,CH); 7.2-7.4 (m,8H,arom.); 8.0 (d,2H,arom.).	21.8 (CH_3); 37.3 (S-CH_2-C); 49.7 (S-CH_2-N); 72.4 (CH); 79.8 (C sp.); 162.7,167.9 (2 CO).	1650 1795	402 261 219
7 a	2.1 (s,3H,CH_3); 2.75-3.1 (dd,2H,S-CH_2-C); 4.45 (dd,2H,S-CH_2N); 5.4 (s,1H,CH);7.3-7.6 (m,8H,arom.); 8.0 (d,2H,arom.).	22.9 (CH_3); 33.1 (S-CH_2-C); 50.4 (S-CH_2-N); 67.1 (CH); 79.4 (C sp.); 164.8,168.4 (2 CO).	1670 1805	402 261 219
6 b	3.1-3.5 (dd,2H,S-CH_2-C); 4.0-4.3 (dd,2H, S-CH_2N); 5.2 (s,1H,CH);6.65 (d,2H,arom); 7.2-7.7 (m,11H,arom.); 8.1 (d,2H,arom.).	----------	1650 1805	464 323 219
7 b	2.7-3.15 (dd,2H,S-CH_2-C); 4.45 (dd,2H, S-CH_2-N); 5.6 (s,1H,CH); 7.3-7.7 (m,13H, aromatics); 8.1 (d,2H,aromatics).	34.6 (S-CH_2-C); 52.8 (S-CH_2-N); 66.2 (CH); 79.7 (C sp.); 164.9, 168.8 (2 CO).	1695 1795	464 323 219
8 b	4.3 (s,2H,S-CH_2-C); 5.2 (s,2H,S-CH_2-N); 7.3-7.7 (m,10H,aromatics).	26.6 (S-CH_2-C); 46.4 (S-CH_2-N).	710 1600	278
6 c	3.1-3.5 (dd,2H,S-CH_2-C);3.75(s,3H,OCH_3); 4.05-4.3 (dd,2H,S-CH_2N); 5.2 (s,1H,CH); 6.7 (m,4H,arom.); 7.25-7.7 (m,8H,arom.); 8.1 (d,2H,aromatics).	37.4 (S-CH_2-C); 51.3 (S-CH_2-N); 55.2 (OCH_3); 72.1 (CH); 79.7 (C sp.); 160.9,162.7 (2 CO).	1635 1795	494 353 219
7 c	2.70-3.15 (dd,2H,S-CH_2-C); 3.85 (s,3H, OCH_3); 4.45-4.55 (dd,2H,S-CH_2-N); 5.6 (s,1H,CH); 6.9 (d,2H,arom.); 7.3-7.7 (m,10H,aromatics); 8.05 (d,2H,aromatics).	34.7 (S-CH_2-C); 52.9 (S-CH_2-N); 55.4 (OCH_3); 66.0 (CH); 79.8 (C sp.); 165.2,168.5 (2 CO).	1645 1795	494 353 219
8 c	3.85 (s,3H,OCH_3); 4.3 (s,2H,S-CH_2-C); 5.15(s,2H,S-CH_2-N);6.95-7.75(m,9H,arom.)	26.6 (S-CH_2-C); 46.3 (S-CH_2-N); 55.2 (OCH_3).	715 1600	308

REFERENCES AND NOTES

1. Dalla Croce,P.; Gariboldi,P.L.; La Rosa,C. *J. Heter. Chem.* **1987**, *24*, 1793.
2. Ballabio,M.; Dalla Croce,P.; Gelmi,M.L.; La Rosa,C. *J. Chem. Soc. Perkin Trans.2* **1988**, 423.
3. Dalla Croce,P.; La Rosa,C. *Heterocycles* **1988**, *27*, 2825.
4. Consonni,R.; Dalla Croce,P.; Ferraccioli,R.; La Rosa,C. *J. Chem. Res. (S)* **1991**, 188.
5. Dalla Croce,P.; Ferraccioli,R.; La Rosa,C.; Pilati,T. *J. Chem. Soc. Perkin Trans. 2* **1993**, 1511 and references therein.
6. Kane,J.M. *J. Org. Chem.* **1980**, *45*, 5396.
7. Nagasawa,H.T.; Goon,D.J.W.; Shirota,F.N. *J. Heter. Chem.* **1981**, *18*, 1047.
8. Anderson,W.K.; Mach,R.H. *J. Med. Chem.* **1987**, *30*, 2109.
9. Ladurée,D.; Lancelot,J.C.; Robba,M.; Chenu,E.; Mathé,G. *J. Med. Chem.* **1989**, *32*, 456.
10. a) Fabre,J.L.; Farge,D.; James,C.; Lavé,D. *Tetrahedron Lett.* **1985**, *26*, 5447. b) Eur.Pat. 147,317 **1985**, C.A. 103:160500 m. c) Fr.Pat. 2,539,417 **1984**, C.A. 102:45932w.
11. Dalla Croce,P.; Ferraccioli,R.;La Rosa,C. *J. Chem. Soc. Perkin Trans.1* **1994**, 2499.
12. a) Cyclodehydrations accomplished with acetic anhydride gave mainly acetanilide with imine **(2)** and a complex mixture with imine **(3)** with very low yields in products **(4-8)**. Other dehydration procedures (Ph_3P/CCl_4/TEA; $SOCl_2$/Py/dioxane) were unsuccessful. b) Beside the products indicated in Scheme 2 it was always isolated a side product deriving from the reaction between **(1)** and the dicyclohexylcarbodiimide, the N-[(3-acyl-thiazolidin-4-yl)carbonyl]-N,N'-dicyclohexyl urea. This product, that was isolated with yields ranging from 25 (R=C_6H_5) to 60% (R=CH_3), is the only formed when we used the N-formyl-(R)-thiazolidine- 4-carboxylic acid as starting material (**1**, R=H). Attempts to carry out the cyclodehydration step at lower temperature (0-5° C) did not lead to cycloaddition products probably owing to the non-formation of **(α)** : Davies,J.S.;Mohammed,A.K. *J. Chem. Soc.Perkin Trans.1* **1981**,2982.
13. a)Huisgen,R.;Funke,E.;Schaefer,F.C.;Knorr,R.*Angew.Chem.Int.Ed.***1967**,*6*,3 67. b) Funke,E.;Huisgen, R.*Chem.Ber.***1971**,*104*,3222.
14. Sheehan,J.C.;Yahg,D.D.H.*J.Am.Chem.Soc.***1958**,*80*,1158.
15. This first step was supposed to occur in the thermal rearrangement of monocyclic 3-acyl-amino-β-lactams : Bird,C.W. *Tetrahedron* **1966**, *22*, 2489 and references therein.
16. Ratner,S.; Clarke,H.T. *J. Am. Chem. Soc.* **1937**, *59*, 200.
17. Neher,R.; Wettstein,A.; Miescher,K. *Helv. Chim. Acta* **1946**, *29*, 1815.
18. Bigelow,L.A.; Eatough,H. *Org. Synth. Coll. Vol.I* **1946**, 80.
19. Jennings,W.B.; Lovely,C.J. *Tetrahedron Lett.* **1988**, *29*, 3725.
20. Albrecht,R.; Kresze,G.; Mlakar,B. *Chem. Ber.* **1964**, *97*, 483.

(*Received in UK* 3 *May* 1995; *revised* 3 *July* 1995; *accepted* 7 *July* 1995)