Reaction of 2,4-Diphenyl-4,5-dihydro-1,3-oxazol-5-one with 4-Phenyl-*N*-tosyl-1-azabuta-1,3-diene: C=C *versus* C=N Double Bond Addition

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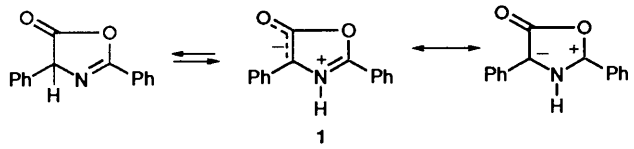
The reaction of 2,4-diphenyl-4,5-dihydro-1,3-oxazol-5-one **1** with 4-phenyl-*N*-tosyl-1-azabuta-1,3-diene **2** has been investigated using different experimental conditions. Whereas at room temperature the kinetically controlled Michael adduct **3** was isolated, at 110 °C products **4**, **5**, **6** and **7** were obtained. Their formation is explained according to 1,3-dipolar cycloaddition and nucleophilic addition, both related to the dual nature of the azlactone **1**. The reactivity of azlactone **1** towards imine **2**, which is different from that observed with *N*-alkyl- and *N*-aryl-1-azabuta-1,3-dienes, is explained on the basis of the electronic effects exerted by the tosyl substituent.

Simple and α,β-unsaturated *N*-phenylsulfonyl substituted imines have recently been used as precursors of heterocycles. Pyrrolidines and tetrahydropyridines can be obtained by treating *N*-phenylsulfonyl-1-azabuta-1,3-dienes with (trimethylenemethane)palladium complexes[1] and with vinyl ethers,[2] respectively. Moreover, azetidines[3] can be prepared from simple imines and enol ethers.

In connection with our work concerning the use of 1,3-oxazolium 5-oxides in heterocyclic synthesis,[4] we observed that simple and α,β-unsaturated *N*-phenylsulfonyl imines underwent a 1,3-dipolar cycloaddition with 1-methyl-1,3-oxazolium 5-oxides, commonly known as Münchnones, leading in the first case to imidazole derivatives[4c] and in the second, to a mixture of imidazole and pyrrole derivatives.[4d] In contrast, it is known that simple and α,β-unsaturated imines, bearing phenyl or alkyl groups on the nitrogen, react at 80 °C with mesoionic 1,3-oxazolium 5-oxides giving β-lactams through a [2 + 2] cycloaddition.[4d,5] Our results provide new opportunities for the synthesis of nitrogen heterocyclic systems because they indicate that the reaction between imines and Münchnones can be selectively directed towards five-membered heterocycles by the use of *N*-phenylsulfonyl substituted imines.

In order to study further the general reactivity of *N*-tosyl-1-azabuta-1,3-dienes we have studied the reaction of 4-phenyl-*N*-tosyl-1-azabuta-1,3-diene **2** with the azlactone 2,4-diphenyl-4,5-dihydro-1,3-oxazol-5-one **1**, chosen on the basis of its dual chemical behaviour, as a nucleophile and 1,3-dipole[6] (Scheme 1).



1

Scheme 1

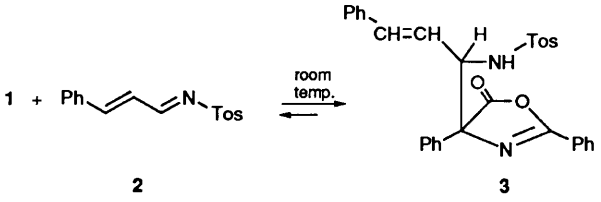
Results and Discussion

The reaction of the imine **2** with azlactone **1** was highly dependent on the experimental conditions. The reaction of **1** with **2** as a suspension in toluene at room temperature under a nitrogen atmosphere for 48 h produced 79% of a solid material with a wide melting range. Through recrystallisation, we obtained two compounds **A** and **B**, which had m.p.s of 128–130 and 153–155 °C, respectively. The analytical properties and spectral data, reported in the Experimental section, are in agreement with the structure **3**. The IR spectra of each adduct showed bands indicating the presence of an NH, an azlactone ring and a carbon–carbon double bond. The 300 MHz ^{1}H NMR spectra of each diastereoisomer showed four sets of signals corresponding to four protons in the range δ 4.0–6.0 for **A** and δ 4.0–6.5 for **B**. The chemical shifts and the coupling constants, confirmed by spin–spin decoupling experiments, showed that these sets of signals belong to the imine framework.†

The ratio **A**:**B** present in the crude precipitate was determined to be 4:1 on the basis of the ^{1}H NMR analysis by evaluating the integration of the olefinic proton at δ 5.9 for compound **A** with respect to the corresponding one at δ 6.5 for compound **B**.

When the reaction of the azlactone **1** with the imine **2** was carried out in a more concentrated toluene suspension, it was observed that after 3 h only the diastereoisomer **3A** was formed in 72% yield.

The formation of the diastereoisomeric adducts **3A** and **B** was due to a Michael addition of the C-4 azlactone carbon to the C=N double bond[7] (Scheme 2). Recent studies on charge



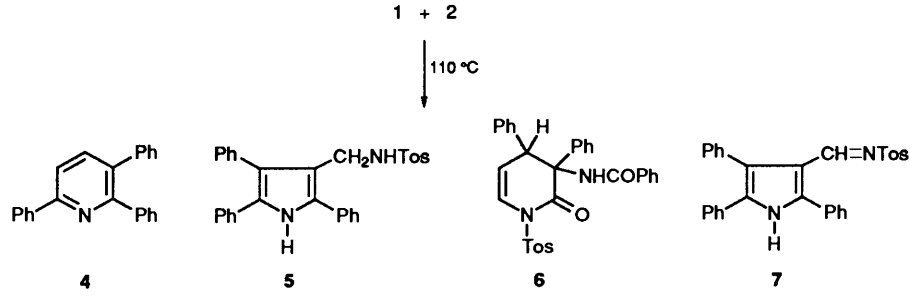
2 **3**

Scheme 2 Tos = *p*-$MeC_6H_4SO_2$

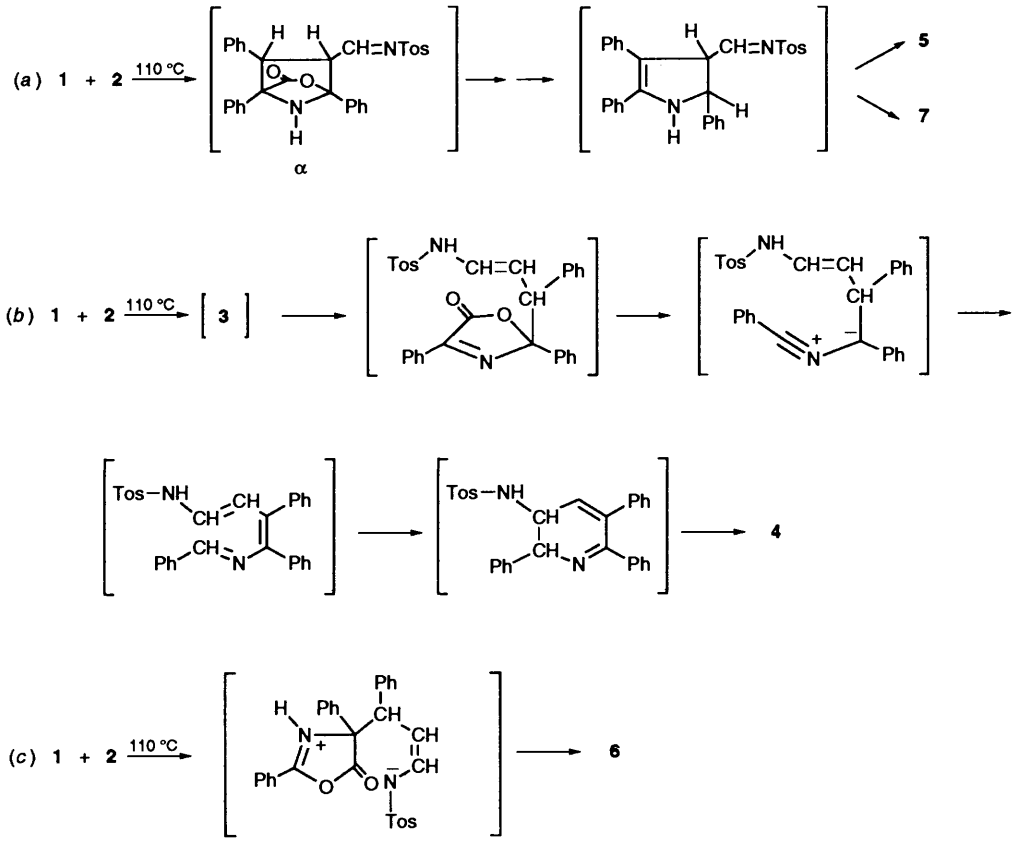
distribution relevant to the parent 1,3-oxazol-5-one, indicate that the C-4 and the exocyclic oxygen are the most negatively charged atoms in the molecule.[8]

Adducts **A** and **B** were very unstable in solution. The ^{1}H NMR analysis of compound **A** in $CDCl_3$ solution at room temperature showed that after 24 h only 50% of the adduct was present and after 48 h it had totally disappeared, and only the ^{1}H NMR signals relevant to the starting materials were present: the significant ones for **2** were those at δ 7.09 (1 H, dd, *J* 16 and 9, PhCH=C*H*) and 8.84 (1 H, d, *J* 9, CH=N); and for **1** the signal at δ 5.5 relevant to 4-H.

† On the basis of our ^{1}H NMR data we were not able to assign the relative configuration of the two stereogenic centres of each diastereoisomer.



Scheme 3



Scheme 4

The diastereoisomeric adducts **3A** and **B** immediately dissociated into the precursors azlactone **1** and imine **2**, both in acidic (2 mol dm^{-3} HCL, THF, room temp.) and basic (1 mol dm^{-3} NaOH, THF, room temp.) media. In order to avoid their dissociation we attempted to alkylate the tosylamino group with diazomethane at temperatures ranging from −5 to −10 °C. The ^1H NMR spectrum of the crude reaction product showed the presence of signals at δ 7.09 and 8.84 diagnostic of compound **2** and an absence of signals in the range δ 4.0–6.5. A further unsuccessful attempt was made using neutral silica gel as a catalyst.[9] This behaviour stopped us from carrying out any further chemical investigation on these adducts.

The reaction of the azlactone **1** with the imine **2** in toluene at 110 °C under a nitrogen atmosphere for 3 h gave a mixture of four products, which were separated by column chromatography and further purified by crystallization. Analytical, physical and spectroscopic data, relevant to each isolated compound are reported in the Experimental section. They fully agree with the structures shown in Scheme 3: pyridine **4** (33%), pyrrole **5** (15%), pyridinone **6** (8%) and pyrrole **7** (5%). In addition to these compounds, toluene-*p*-sulfonamide was also isolated in a stoichiometric amount with respect to compound **4**.

To explain the formation of this product mixture we hypothesize that the reaction occurs according to two mechanisms. 1,3-Dipolar cycloaddition of the azlactone **1** to the C=C double bond of the imine **2** accounts for the formation of pyrrole derivatives **5** and **7**; and a nucleophilic addition of C-4 of the azlactone to C-2 and C-4 of the imine accounts for compounds **4** and **6**, respectively.

In the first case, as shown in Scheme 4(*a*), compound **1** in its mesoionic form reacted at the C=C double bond giving a 1,3-dipolar cycloadduct **α**.[6] Upon losing carbon dioxide it led to dihydropyrrole derivative intermediate which underwent either a rearrangement leading to **5** or an oxidation producing **7**.

To explain the formation of compound **4** [see Scheme 4(*b*)] we hypothesize the initial formation of compound **3**. The allylic substituent at C-4 allows the 4,5-dihydrooxazolone **3** to undergo the Cope rearrangement[10] at 110 °C leading to the corresponding 2,5-dihydrooxazolone. On further heating, the 2,5-dihydrooxazolone easily lost carbon dioxide giving a nitrile ylide intermediate.[11] This underwent a rearrangement producing a dihydropyridine which aromatized *via* toluene-*p*-sulfonamide elimination. We proved that the adducts **3** were initially involved in the pyridine derivative formation by heating the adduct **3A** in toluene for 3 h at 110 °C under a nitrogen

atmosphere. As predicted, the reaction gave compound **4** (58%) and the pyrrole derivative **5** (4%).

To account for the pyridin-2-one **6** formation, three different hypotheses can be taken in consideration.[12c] The first one, which involves initial imine nitrogen attack on the azlactone carbonyl group could be ruled out in our case because the nitrogen is not a good nucleophile. Likewise the well-documented reactivity of **2** towards electron-rich dienophiles[2,3] seemed to rule out the second possibility based on the [4 + 2] cycloaddition between the imine **2** and the valence tautomeric ketene intermediate of the azlactone. However, the most likely explanation seemed to involve an initial attack of the negatively charged carbon atom of **1** on C-4 of **2**, followed by a six-membered ring closure [Scheme (*c*)].

Under basic conditions (triethylamine at 0 °C) the reaction of the azlactone **1** with the imine **2** became highly selective giving pyridinone **6** in quantitative yield. From the total absence of adducts **3** from azlactone C-4 addition to the C=N double bond, it was deduced that they were kinetically controlled products. Since the adducts **3** cannot be transformed into pyridine derivative because of the low temperature, under basic conditions they dissociated back into the starting materials **1** and **2** which finally gave compound **6**. To check this hypothesis, we carried out a reaction in which the diastereoisomeric adduct **3A** was treated with a stoichiometric amount of triethylamine at 0 °C, in toluene solution and we obtained compound **6** (85%) as the only product.

On the basis of the addition products isolated and their yields it is clear that two electrophilic sites, namely C-2 and C-4, are present in imine **2** and that C-2 is the most electrophilic. As the LUMO coefficient values (C-2 = −0.58 and C-4 = 0.53 eV) of the parent *N*-phenylsulfonyl-1-azabuta-1,3-diene[2] taken as a reference for **2** are almost the same, the preferred imine C-2 addition is due to the electrostatic interaction between the two reactants. As the tosyl substituent polarizes the C=N more than the C=C double bond, the polar interaction between the imine system and the highly polarized azlactone favours the C-2 addition product.

As a comparison with our results it is worth noting that *N*-alkyl and *N*-aryl α,β-unsaturated imines react with azlactone **1** giving only the corresponding pyridin-2-one derivatives.[12] It emerges that the 1,3-dipolar cycloaddition between **1** and the C=C double bond of **2**, giving pyrrole derivatives **5** and **7** is the most substantial difference in the reactivity between *N*-alkyl and *N*-aryl, and *N*-tosyl imines. This result confirms the capability of *N*-tosylimines to undergo 1,3-dipolar cycloadditions. Compound **1**, resembling in its mesoionic form a Münchnone system, is expected to be analogously reactive with electron-deficient dipolarophiles.[6] The electron-withdrawing tosyl substituent decreases the LUMO energy levels of 1-azabuta-1,3-dienes, as a comparison between the FMO energy levels of unsubstituted 1-azabuta-1,3-diene (HOMO: −10.1; LUMO: 0.4 eV) and the *N*-phenylsulfonyl derivative (HOMO: −11.1; LUMO: −0.9 eV)[2] indicates. On this basis we can predict that compound **2** fulfils the requirements of electron-deficiency more than *N*-alkyl, *N*-aryl substituted imines.

As previously reported, the mesoionic 1-methyl-1,3-oxazolium 5-oxide underwent 1,3-dipolar cycloaddition with both C=C and C=N double bonds of 4-phenyl-*N*-phenylsulfonyl-1-azabuta-1,3-dienes leading to pyrrole and imidazole derivatives, respectively.[4d] However, the isolation of the dihydrooxazalone **3** at room temperature and the pyridine **4** at 110 °C confirms that a concerted 1,3-dipolar cycloaddition between the azlactone **1**, in its mesoionic form, and the C=N double bond of the imine **2** does not occur at all. This process would give a primary bicyclic adduct which *via* carbon dioxide and toluene-*p*-sulfinic acid elimination[4d] would lead to the 2,5-diphenyl-4-styrylimidazole, which was never detected. The lack of the imidazole derivative indicates that the zwitterionic intermediate involved in the first step undergoes an intramolecular proton transfer to dihydrooxazolone **3** faster than the ring closure to the bicyclic adduct.

Experimental

M.p.s were measured with a Buchi apparatus and are uncorrected. IR spectra were obtained with a Perkin-Elmer 298 spectrophotometer. ^{1}H NMR spectra were recorded on Bruker WP 80-SY and Bruker AC 300 spectrometers. All chemical shifts are expressed in δ values from tetramethylsilane as the reference. *J* Values are expressed in Hz. Positive ion FAB MS spectra were determined on a VG Analytical 7070 EQ mass spectrometer with a VG Analytical 11/250 data system attached. Compounds **2**[13] and *N*-benzoylphenylglycine as well as the corresponding oxazol-5-one **1** were prepared according to the reported procedure.[14] Solvents were dried according to standard procedures.

Reaction of 2,4-*Diphenyl*-4,5-*dihydro*-1,3-*oxazol*-5-*one* **1** *with* N-(3-*Phenylprop*-2-*enylidene*)*toluene*-p-*sulfonamide* **2** *at Room Temperature: Preparation of the Diastereoisomers of* 2,4-*Diphenyl*-2-[3-*phenyl*-1-(*toluene*-p-*sulfonamido*)*prop*-2-*enyl*]-4,5-*dihydro*-1,3-*oxazol*-5-*one* **3**.—To a yellow suspension of oxazolone **1** (0.54 g, 2.3 mmol) in dry toluene (10 cm^3) was added sulfonamide **2** (0.65 g, 2.3 mmol). The reaction mixture was stirred under a nitrogen atmosphere at room temp. for 48 h. During this time a precipitate formed at the same time as the discolouration of the solution occurred. The mixture was filtered and the filtrate was concentrated to give a white solid (0.95 g, 79%). A sample of the crude residue was dissolved in ethyl acetate at 40 °C and kept at −20 °C for several hours to give a solid which was the diastereoisomer **3B**, m.p. 153–155 °C (Found: C, 71.25; H, 5.0; N, 5.4. $C_{31}H_{26}N_2O_4S$ requires: C, 71.1; H, 5.1; N, 5.3%); ν_{max}(Nujol)/cm^{-1} 3300 (NH), 1815 (CO) and 1660 (C=C); δ_H(300 MHz; $CDCl_3$) 2.2 (3 H, s, Me), 4.7 (1 H, t, *J* 8, C*H*NH), 5.13 (1 H, d, *J* 8, NH), 5.68 (1 H, dd, *J* 8 and 16, =CCHN), 6.5 (1 H, d, *J* 16, PhC*H*=), 6.98–7.68 (17 H, m, ArH) and 8.02 (2 H, d, *J* 7, ArH); FAB *m*/*z* 522 (M^+). To the mother liquor was added a quantity of diisopropyl alcohol (ethyl acetate–diisopropyl alcohol, 9:1) and the solution was kept at −20 °C for several hours to give the diastereoisomer **3A**, m.p. 128–130 °C (Found: C, 71.3; H, 5.0; N, 5.2. $C_{31}H_{26}N_2O_4S$ requires: C, 71.1; H, 5.1; N, 5.3%); ν_{max}(Nujol)/cm^{-1} 3240, 1820, 1840 and 1650; δ_H(300 MHz; $CDCl_3$) 2.25 (3 H, s, Me), 4.65–4.75 (2 H, m, CHN, NH), 5.71 (1 H, dd, *J* 15.9 and 7.06, =CCHN), 5.90 (1 H, d, *J* 15.9, PhC*H*=), 6.85–7.70 (17 H, m, ArH) and 8.05 (2 H, d, *J* 7.4, ArH); FAB *m*/*z* 522 (M^+).

Preparation of the Diastereoisomer **3A**.—To a suspension of oxazolone **1** (1 g, 4.2 mmol) in dry toluene (12 cm^3) was added sulfonamide **2** (1.2 g, 4.2 mmol). The reaction mixture was stirred under a nitrogen atmosphere at room temperature for 3 h during which time a precipitate formed. The mixture was filtered and the filtrate was concentrated to give a pale yellow solid (1.8 g, 72%), which was shown by ^{1}H NMR analysis to be diastereoisomer **3A**. When the filtrate was evaporated to dryness, the mother liquor gave a crude oily residue. The ^{1}H NMR spectrum showed only traces of **A** and the absence of **B**.

Reaction between Oxazolone **1** *and Sulfonamide* **2** *at* 110 °C.—A suspension of oxazolone **1** (1.66 g, 7.5 mmol) and sulfonamide **2** (2.0 g, 7.0 mmol) in toluene (50 cm^3) was heated at 110 °C for 3 h. During this time CO_2 evolution was observed. The reaction mixture was evaporated and the residue, which was shown by TLC analysis (toluene–ethyl acetate, 9:1) to be a complex

mixture, was separated by chromatography on silica gel (toluene–ethyl acetate, 9:1) to give five components described here according to the order of elution.

2,3,6-Triphenylpyridine **4** (0.7 g, 33%), m.p. 112 °C (from diisopropyl alcohol) (lit.,[15] 115 °C); 2,4,5-triphenyl-3-(toluene-*p*-sulfonamidomethyl)pyrrole **5** (0.5 g, 15%), m.p. 238–239 °C (from diisopropyl alcohol) (Found: C, 75.3; H, 5.5; N, 5.85. $C_{30}H_{26}N_2O_2S$ requires C, 75.45; H, 5.6; N, 6.05%); ν_{max} (Nujol)/cm^{-1} 3310 and 3200; δ_H(300 MHz; $CDCl_3$) 2.45 (3 H, s, Me), 4.09 (2 H, d, *J* 4.9, CH_2N), 4.2 (1 H, t, *J* 4.9, NH), 7.1–7.5 (19 H, m, ArH) and 8.4 (1 H, br s, NH); 3-benzamido-3,4-diphenyl-*N*-(*p*-tolylsulfonyl-1,2,3,4-tetrahydropyridin-2-one **6** (0.3 g, 8%), m.p. 200–201 °C (from ethyl alcohol) (Found: C, 71.25; H, 5.0; N, 5.4. $C_{31}H_{26}N_2O_4S$ requires: C, 71.1; H, 5.1; N, 5.55%); ν_{max}(Nujol)/cm^{-1} 3240, 1710, 1675 and 1650; δ_H(300 MHz; $CDCl_3$) 2.5 (3 H, s, Me), 5.3 (1 H, d, *J* 7, PhC*H*), 5.8 (1 H, t, *J* 7, CC*H*=CH), 6.8–7.5 (19 H, m, ArH), CH=C*H*N, NH) and 7.9 (2 H, d, *J* 8, ArH); δ_C(300 MHz; $CDCl_3$) 21.73 (Me), 42.44 (C-4), 65.73 (C-3), 112.82 (C-5), 122.98 (C-6), 165.76 (CO) and 168.25 (CO); FAB *m/z* 523 (M + 1); *N*-(2,4,5-triphenylpyrrol-3-ylmethylene)toluene-*p*-sulfonamide **7** (0.17, 5%), m.p. 188–189 °C (from diisopropyl alcohol) (Found: C, 75.6; H, 5.1; N, 5.9. $C_{30}H_{24}N_2O_2S$ requires C, 75.45; H, 5.2; N, 6.0%); ν_{max} (Nujol)/cm^{-1} 3220 and 1590; δ_H(80 MHz; $CDCl_3$) 2.35 (3 H, s, Me), 7.1–7.6 (19 H, m, ArH), 8.6 (1 H, s, NH) and 8.85 (1 H, s, CH=); toluene-*p*-sulfonamide (0.39 g, a stoichiometric amount with respect to the compound **5**).

Reaction between Oxazolone **1** *and Sulfonamide* **2** *in the Presence of* Et_3N*: Synthesis of Pyridinone* **6**.—To a suspension of oxazolone **1** (0.3 g, 1.3 mmol) in toluene (20 cm^3), at 0 °C, was added dropwise a solution of sulfonamide **2** (0.370 g, 1.3 mmol) and triethylamine (0.13 g, 1.3 mmol) in toluene (10 cm^3). At the end of the addition, the solution was allowed to warm to room temp. and then washed with brine (15 cm^3). The organic phase was separated, dried with Na_2SO_4 and then evaporated. The residue was purified by chromatography (toluene–ethyl acetate, 8:2) to give compound **6** (0.54 g, 80%) as the only reaction product.

Transformation of Oxazolone **3** *into Pyridine* **4** *and Pyrrole* **5** *at* 110 C.—A suspension of adduct **3A** (1 g, 1.9 mmol) in toluene (30 cm^3) was heated at 110 °C until the evolution of CO_2 ceased (3 h). The reaction mixture was then evaporated to dryness. TLC analysis (toluene–ethyl acetate 9:1) showed that the residue obtained consisted of three components which were separated by chromatography (toluene–ethyl acetate, 9:1) and then crystallised. On the basis of analytical and spectroscopic data, the first component eluted was identified as compound **4** (0.340 g, 58%), the second one as compound **5** (0.04 g, 4%), the third one as toluene-*p*-sulfonamide (0.15 g, a stoichiometric amount with respect to compound **4**).

Reaction of Diastereoisomer **3A** *with* Et_3N*: Synthesis of Pyridinone* **6**.—To a solution of adduct **3A** (0.5 g, 0.96 mmol) in dry toluene (8 cm^3), at 0 °C under a nitrogen atmosphere, was added dry triethylamine (0.120 g, 0.15 cm^3, 1.1 mmol) in dry toluene (3 cm^3). After the addition, the reaction mixture was left at room temperature for 15 min. TLC analysis (toluene–ethyl acetate, 9:1) of the solution showed the formation of a product having the same R_f as compound **6**. The solution was washed with water (5 cm^3). The organic layer was separated, dried with Na_2SO_4 and then evaporated. A solid was obtained (0.420 g, 85%) whose physical and spectroscopic properties were coincident with those reported for compound **6**.

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