Monatshefte für Chemie 134, 1105–1111 (2003)
DOI 10.1007/s00706-003-0020-6

Monatshefte für Chemie
Chemical Monthly
Printed in Austria

Comparison of Conventional and Microwave Assisted Reactions Giving Aromatic Oxazolidines

Florian Diwischek, Eberhard Heller, and **Ulrike Holzgrabe***

Institute of Pharmacy and Food Chemistry, Am Hubland, University of Würzburg, D-97074 Würzburg, Germany

Received December 19, 2002; accepted December 23, 2002
Published online June 2, 2003

Summary. By reaction of aromatic aldehydes with (-)-ephedrine aromatic 1,3-oxazolidines can be obtained. The reaction was carried out either at conventional conditions or by microwave heating. The different diastereomeric ratios were determined by means of ^{1}H NMR spectroscopy.

Keywords. Oxazolidines; Microwave chemistry; Diastereomeric ratio.

Introduction

Microwaves are increasingly used in chemical synthesis. Compared to conventional heating, microwave dielectric heating exhibits the following thermal differences [1]: The introduction of microwave energy into a reaction mixture which contains at least one compound capable of absorbing microwaves strongly, leads to much higher heating rates. Microwave energy is introduced into a chemical reaction immediately and without direct contact between energy source and solution. In addition, chemicals do not interact equally with the microwave frequency applied. This may lead to a different temperature profile of the reaction and, therefore, may result in an alternative distribution of products compared to conventional heating. Application of closed reaction tubes allows heating up to temperatures much higher than the conventional boiling point of the solvent used. The higher temperature may lead to a large acceleration of reactions. Even though non-thermal effects (effects specially inherent to microwaves and not caused by different temperature regimes) were initially claimed, it now seems to be generally accepted that the different temperature regime caused by microwave heating is the main responsible factor to any acceleration observed [2]. However, the subject of non-thermal effects in microwave heating is still intensively discussed [3, 4].

* Corresponding author. E-mail: u.holzgrabe@pharmazie.uni-wuerzburg.de

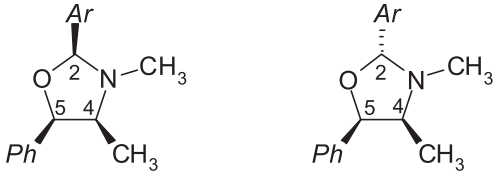
As a test reaction for the comparison of a conventional reaction setup with a microwave-assisted one, the condensation of carbonyl compounds with amino alcohols giving diastereomeric oxazolidines was studied.

Results and Discussion

Conventional synthesis routes to oxazolidines have been reported often in literature [5–8]. *Neelakantan* [9] has found a remarkable diastereoselective oxazolidine formation by conversion of aromatic aldehydes with (-)-ephedrine. *Neelakantan's* stereochemical assignments [10] have been a matter of discussion [11]. In contrast to *Neelakantan* the resulting major isomer **a** represents the (*S*)-configuration at C2 and the second diastereomer **b** (2*R*) does not exceed 10% in the isolated product mixture (see Fig. 1) [12]. The resulting diastereomers of the oxazolidines can be used as chiral auxiliaries for stereoselective synthesis [12–14].

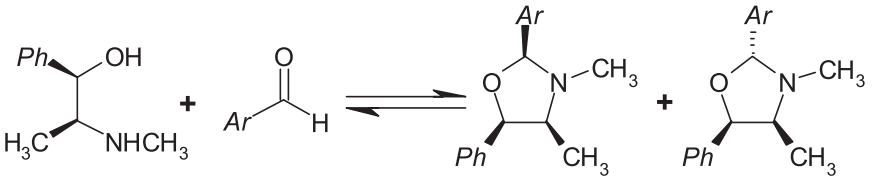
Kuhnert et al. have investigated the formation of 1,3-oxazolidines with focused microwaves under thermodynamic control [15]. In contrast to their investigation we compared the diastereomeric ratios of the classical way of oxazolidine synthesis with the one in which we used microwave heating for different periods of times and at different temperatures.

The reaction mechanism is characterized by the initial formation of an iminium ion formed by ephedrine and the aldehyde, which reacts to the oxazolidine by an intramolecular addition of the hydroxy group onto the iminium ion. Studying the oxazolidine formation with different aromatic aldehydes *Agami et al*. [16] have observed for benzaldehyde and 4-methoxybenzaldehyde the immediate formation of isomer **a** as the major product. For 4-cyano- and 4-nitrobenzaldehydes a ratio of **a**:**b** = 1:1 was found by means of NMR spectroscopy at the beginning of the reaction in chloroform. However, at the end of the reaction isomer **a** was the major product. Therefore, isomer **a** is supposed to be the thermodynamically controlled and isomer **b** the kinetically controlled product. This is supported by results obtained by *Beckett* [17] who has investigated the reaction of (-)-ephedrine and acetaldehyde and has also observed the formation of both diastereomers at the beginning of the reaction and, subsequently, the conversion of the (2*R*) into the (2*S*) isomer. Performing the reaction of 4-cyano- and 4-nitrobenzaldehyde in methanol, isomer **b** could be only observed at the beginning of the reaction, which also resulted in **a** as the predominating isomer in the end. Finally, even isomer **b** could be isolated ($Ar = 4\text{-}C_6H_4Br$) after the reaction being carried out in aqueous ethanol under carefully controlled conditions [16].



a **b**

Fig. 1. Oxazolidine diastereomers



8 - 14 **1a - 7a** **1b - 7b**

Ar = —(C_6H_4)—*R*

	1/8	**2/9**	**3/10**	**4/11**	**5/12**	**6/13**	**7/14**
R	H	4-CH_3	4-NMe_2	4-OMe	4-NO_2	2-NO_2	2-Cl

Scheme 1. Synthesis of oxazolidines **1**–**7**; conventional method: 25°C, 23–48 h; microwave assisted reaction: 100, 120°C, 5, 10, 30 min

The aim of this investigation was to find out whether differences in the isomeric ratio occur when the reaction is carried out under conventional conditions compared to using microwaves at different temperatures and various periods of time. Benzaldehydes of different substitution pattern (*R* = H, 4-CH_3, 4-N*Me*$_2$, 4-O*Me*, 4-NO_2, 2-NO_2, 2-Cl; **8**–**14**) were considered in this study. The progress of the reaction was monitored by TLC and the isomeric ratio was determined by means of NMR spectroscopy.

In order to evaluate the isomeric ratio the ^{1}H NMR spectra were first assigned. The integral of the signals of H2 of both isomers, which are well separated (*e.g.* $\delta = 4.62$ ppm *versus* 5.26 ppm for **1a** and **1b**) was used as a measure of the content of each isomer in the reaction mixture. In the case of 2-(2-nitrophenyl)-5-phenyl-3,4-dimethyloxazolidine signals of H2 and H5 were overlapping. Thus, the integrals of H4 signals were evaluated to measure the diastereomeric ratio.

For each aromatic aldehyde at least one conventional and four microwave experiments were executed. The results are reported in Table 1. The most remarkable finding is the shortening of the reaction time from 1–2 days to minutes, which was observed for all examples. Close inspection of the data obtained from microwave heating revealed the diastereomeric ratio partially to be dependent on the temperature and the time of reaction.

Concerning the comparison between conventional and microwave reactions it can be pointed out that for the formation of oxazolidines **1**–**4** the same or nearly the same diastereomeric ratios were achieved but in much shorter reaction times in the microwave oven. The corresponding aldehydes are characterised by either an electron donating group (**9**–**11**) or no substitution (**8**).

The most striking differences in the stereoisomeric ratio of the products between the conventional and the microwave assisted experiments were obtained in the case of aromatic aldehydes substituted with electron withdrawing groups. For 2-(4-nitrophenyl)-5-phenyl-3,4-dimethyloxazolidine (**5a**) and (**5b**) a higher selectivity is achieved by the conventional method. Increasing time and temperature in the microwave assisted experiments the isomeric ratio of

Table 1. Isomeric ratio of oxazolidine diastereomers (2*S*,4*S*,5*R*)-(**1a**–**7a**):(2*R*,4*S*,5*R*)-(**1b**–**7b**) in dependance on the kind and time of heating (CW = conventional way; MW = microwave-assisted reaction); the ratio was determined by integration of the ^{1}H NMR spectra; for **5a** and **5b** the integrals of the H4 signals were used, in all other cases the signal of H2

Kind of heating	CW	MW	MW	MW	MW	MW
Time/min	2880	5	10	5	10	30
Temp/°C	25	100	100	120	120	120
1	92:8	89:11	97:3	89:11	97:3	–
2	92:8	92:8	91:9	92:8	92:8	92:8
3	93:7	93:7	93:7	93:7	93:7	93:7
4	93:7	90:10	91:9	90:10	92:8	93:7
5	93:7*	81:19	82:18	83:17	87:13	–
6	84:16	65:35	88:12	77:23	>98:<2	–
7	>98:<2	85:15	83:17	92:8	94:6	–

* Reaction time 1380 min

5a:**5b** increases but does not reach the selectivity obtained conventionally. For 2-(2-nitrophenyl)-5-phenyl-3,4-dimethyloxazolidine (**6a**) and (**6b**) a diastereoselective reaction takes place at 120°C in the microwave oven, whereas the conventional experiment results in an isomeric mixture. For 2-(2-chlorophenyl)-5-phenyl-3,4-dimethyloxazolidine (**7a**) and (**7b**) the result resembles the findings for **5**, a diastereoselective reaction can be observed conventionally. Applying higher temperatures in the microwave oven the diastereoselectivity is enhanced, but again does not reach the conventionally obtained selectivity.

Conclusion

The experiments revealed a strong effect on the formation of oxazolidines when using microwaves. A tremendous shortening of reaction time was observed in all cases. The strongest differences in diastereomeric ratios between conventional method and the microwave assisted reaction could be found when using aromatic aldehydes substituted with electron withdrawing groups.

Experimental

Microwave reactions were carried out in a Milestone MLS-Ethos 1600 using a quartz glass vessel (length 15 cm, diameter 2 cm) equipped with a 20 bar excess pressure valve, which was procured from Mikrowellen-Laborsysteme (MWS).

^{1}H NMR (400.13 MHz) and ^{13}C NMR (100.61 MHz) spectra were recorded on a Bruker Avance 400 MHz spectrometer, equipped with XWIN-NMR software running on a Microsoft Windows PC. $CDCl_3$ was applied as solvent and the center of the signal of $CDCl_3$ was used as an internal reference. Thin layer chromatography was performed using aluminium oxide 60 F_{254} neutral plastic sheets.

General

The following commercially available aromatic aldehydes were used: benzaldehyde (**8**), 4-methylbenzaldehyde (**9**), 4-(dimethylamino)benzaldehyde (**10**), 4-methoxy-benzaldehyde (**11**), 4-nitrobenzaldehyde (**12**), 2-nitrobenzaldehyde (**13**), 2-chloro-benzaldehyde (**14**).

Preparation of Oxazolidines via the Conventional Route [16]

The aromatic aldehyde (1.2 mmol) and 1.2 mmol of (-)-ephedrine were dissolved in 15 cm^3 of $CHCl_3$ and 1.0 g of molecular sieve (4 Å) was added. The solution was stirred over a period of 23–48 h at 25°C. To isolate the oxazolidine the mixture was filtered and the solvent was evaporated *in vacuo*.

Preparation of Oxazolidines via Microwave Heating

The aromatic aldehyde (1.2 mmol), 1.2 mmol of (-)-ephedrine, 15 cm^3 of $CHCl_3$, and molecular sieve 4 Å (1.0 g) were added to a sealed tube and placed in the microwave oven. The solution was heated in two minutes to a temperature of 100 or 120°C, kept at this temperature for 5, 10, or 30 min and then cooled down to 40°C within 10 min by additional cooling. The mixture was filtered and the solvent was removed *in vacuo* to give the oxazolidine.

3,4-Dimethyl-2,5-diphenyloxazolidine (**1**)

(2S,4S,5R)-3,4-Dimethyl-2,5-diphenyloxazolidine (**1a**, $C_{17}H_{19}NO$): ^{1}H NMR: $\delta = 7.62–7.14$ (m, $10H_{arom}$), 5.08 (d, $J = 8.1$ Hz, H5), 4.62 (s, H2), 2.86–2.93 (m, H4), 2.12 (s, NCH_3), 0.72 (d, $J = 6.6$ Hz, $CHCH_3$) ppm; ^{13}C NMR: $\delta = 139.8$, 138.1, 129.1, 128.5, 128.3, 128.1, 127.8, 126.0 (C_{arom}), 98.9 (C2), 82.5 (C5), 64.0 (C4), 35.7 (NCH_3), 15.5 ($CHCH_3$) ppm.

(2R,4S,5R)-3,4-Dimethyl-2,5-diphenyloxazolidine (**1b**, $C_{17}H_{19}NO$): ^{1}H NMR: $\delta = 7.62–7.14$ (m, $10H_{arom}$), 5.51 (d, $J = 5.3$ Hz, H5), 5.26 (s, H2), 3.68–3.60 (m, H4), 2.19 (s, NCH_3), 0.65 (d, $J = 6.6$ Hz, $CHCH_3$) ppm; ^{13}C NMR: $\delta = 141.1$, 138.2, 129.8, 128.0, 127.9, 127.6, 127.1, 126.2 (C_{arom}), 95.3 (C2), 82.5 (C5), 61.7 (C4), 33.5 (NCH_3), 9.0 ($CHCH_3$) ppm.

3,4-Dimethyl-2-(4-methylphenyl)-5-phenyloxazolidine (**2**)

(2S,4S,5R)-3,4-Dimethyl-2-(4-methylphenyl)-5-phenyloxazolidine (**2a**, $C_{18}H_{21}NO$): ^{1}H NMR: $\delta =$ 7.65–7.05 (m, 9H, C_6H_5, $C_6H_4CH_3$), 5.01 (d, $J = 8.1$ Hz, H5), 4.55 (s, H2), 2.86–2.76 (m, H4), 2.26 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, $C_6H_4CH_3$), 2.13–2.05 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, NCH_3), 0.66 (d, $J = 6.6$ Hz, $CHCH_3$) ppm; ^{13}C NMR: $\delta = 139.8$, 139.4, 135.3, 129.1, 128.2, 127.9, 127.7, 126.1 (C_{arom}), 99.2 (C2), 82.8 (C5), 64.4 (C4), 36.2 (NCH_3), 21.8 ($C_6H_4CH_3$), 15.5 ($CHCH_3$) ppm.

(2R,4S,5R)-3,4-Dimethyl-2-(4-methylphenyl)-5-phenyloxazolidine (**2b**, $C_{18}H_{21}NO$): ^{1}H NMR: $\delta = 7.05–7.65$ (m, 9H, C_6H_5, $C_6H_4CH_3$), 5.46 (d, $J = 5.3$ Hz, H5), 5.20 (s, H2), 3.52–3.62 (m, H4), 2.26 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, $C_6H_4CH_3$), 2.05–2.13 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, NCH_3), 0.60 (d, $J = 6.8$ Hz, $CHCH_3$) ppm; ^{13}C NMR: $\delta = 138.4$, 138.2, 136.9, 129.6, 128.3, 127.9, 127.4, 126.1 (C_{arom}), 95.7 (C2), 82.5 (C5), 62.1 (C4), 34.0 (NCH_3), 21.8 ($C_6H_4CH_3$), 9.5 ($CHCH_3$) ppm.

2-[4-(Dimethylamino)phenyl]-3,4-dimethyl-5-phenyloxazolidine (**3**)

(2S,4S,5R)-2-[4-(Dimethylamino)phenyl]-3,4-dimethyl-5-phenyloxazolidine (**3a**, $C_{19}H_{24}N_2O$): ^{1}H NMR: $\delta = 7.79–6.72$ (m, 9H, C_6H_5, $C_6H_4NMe_2$), 5.14 (d, $J = 8.3$ Hz, H5), 4.65 (s, H2), 2.90–3.05 (m, 7H, overlapping hydrogens of (2*S*) and (2*R*) isomers, $N(CH_3)_2$ + overlapping hydrogen H4), 2.19 (s, NCH_3), 0.81 (d, $J = 6.3$ Hz, $CHCH_3$) ppm; ^{13}C NMR: $\delta = 151.3$ ($CNMe_2$), 140.2, 129.2, 128.0, 127.8, 127.4, 125.6, 112.3 (C_{arom}), 98.0 (C2), 81.0 (C5), 62.9 (C4), 39.5 ($N(CH_3)_2$), 34.7 (NCH_3), 14.0 ($CHCH_3$) ppm.

(2R,4S,5R)-2-[4-(Dimethylamino)phenyl]-3,4-dimethyl-5-phenyloxazolidine (**3b**, $C_{19}H_{24}N_2O$): ^{1}H NMR: $\delta = 7.79–6.72$ (m, 9H, C_6H_5, $C_6H_4NMe_2$), 5.59 (d, $J = 5.3$ Hz, H5), 5.30 (s, H2), 3.70–3.78 (m, H4), 3.05–2.90 (m, 6H, overlapping hydrogens of (2*S*) and (2*R*) isomers, $N(CH_3)_2$), 2.28 (s, NCH_3), 0.74 (d, $J = 6.6$ Hz, $CHCH_3$) ppm; ^{13}C NMR: $\delta = 154.3$ ($CNMe_2$), 145.7, 132.0, 128.5, 127.7, 127.0, 125.6, 111.0 (C_{arom}), 98.0 (C2), 81.0 (C5), 60.6 (C4), 39.0 ($N(CH_3)_2$), 32.5 (NCH_3), 7.8 ($CHCH_3$) ppm.

2-(4-Methoxyphenyl)-3,4-dimethyl-5-phenyloxazolidine (**4**)

(2S,4S,5R)-2-(4-Methoxyphenyl)-3,4-dimethyl-5-phenyloxazolidine (**4a**, $C_{18}H_{21}NO_2$): ^{1}H NMR: $\delta = 7.78–6.80$ (m, 9H, C_6H_5, C_6H_4OMe), 5.03 (d, $J = 8.3$ Hz, H5), 4.56 (s, H2), 3.73 (s, OCH_3), 2.89–2.81 (m, H4), 2.07 (s, NCH_3), 0.69 (d, $J = 6.3$ Hz, CHCH_3) ppm; ^{13}C NMR: $\delta = 160.3$ (*C*OMe), 139.8, 131.9, 129.6, 128.0, 127.8, 126.0, 113.6 (C_{arom}), 98.5 (C2), 82.2 (C5), 63.9 (C4), 55.2 (OCH_3), 35.6 (NCH_3), 15.0 (CH*C*H_3) ppm.

(2R,4S,5R)-2-(4-Methoxyphenyl)-3,4-dimethyl-5-phenyloxazolidine (**4b**, $C_{18}H_{21}NO_2$): ^{1}H NMR: $\delta = 7.78–6.80$ (m, 9H, C_6H_5, C_6H_4OMe), 5.49 (d, $J = 5.3$ Hz, H5), 5.32 (s, H2), 3.78 (s, 3H, OCH_3), 2.73–2.68 (m, H4), 2.15 (s, NCH_3), 0.62 (d, $J = 6.8$ Hz, CHCH_3) ppm; ^{13}C NMR: $\delta = 164.5$ (*C*OMe), 141.5, 130.2, 129.8, 128.8, 127.0, 126.1, 114.2 (C_{arom}), 98.4 (C2), 81.8 (C5), 61.5 (C4), 55.5 (OCH_3), 33.3 (NCH_3), 8.7 (CH*C*H_3) ppm.

3,4-Dimethyl-2-(4-nitrophenyl)-5-phenyloxazolidine (**5**)

(2S,4S,5R)-3,4-Dimethyl-2-(4-nitrophenyl)-5-phenyloxazolidine (**5a**, $C_{17}H_{18}N_2O_3$): ^{1}H NMR: $\delta =$ 8.23–7.18 (m, 9H, C_6H_5, $C_6H_4NO_2$), 5.10 (d, $J = 8.4$ Hz, H5), 4.72 (s, H2), 3.01–2.93 (m, H4), 2.14 (s, NCH_3), 0.72 (d, $J = 6.6$ Hz, CHCH_3) ppm; ^{13}C NMR: $\delta = 148.5$ (CNO_2), 145.4, 139.0, 129.2, 128.2, 127.9, 126.1, 123.7 (C_{arom}), 97.3 (C2), 82.9 (C5), 64.0 (C4), 35.8 (NCH_3), 15.0 (CH*C*H_3) ppm.

(2R,4S,5R)-3,4-Dimethyl-2-(4-nitrophenyl)-5-phenyloxazolidine (**5b**, $C_{17}H_{18}N_2O_3$): ^{1}H NMR: $\delta =$ 8.23–7.18 (m, 9H, C_6H_5, $C_6H_4NO_2$), 5.48 (d, $J = 5.3$ Hz, H5), 5.31 (s, H2), 3.68–3.59 (m, H4), 2.20 (s, NCH_3), 0.66 (d, $J = 6.8$ Hz, CHCH_3) ppm; ^{13}C NMR: $\delta = 148.2$ (*C*NO_2), 147.4, 138.5, 130.4, 128.5, 128.0, 127.4, 123.5 (C_{arom}), 94.1 (C2), 82.4 (C5), 61.7 (C4), 33.5 (NCH_3), 9.0 (CH*C*H_3) ppm.

3,4-Dimethyl-2-(2-nitrophenyl)-5-phenyloxazolidine (**6**)

(2S,4S,5R)-3,4-Dimethyl-2-(2-nitrophenyl)-5-phenyloxazolidine (**6a**, $C_{17}H_{18}N_2O_3$): ^{1}H NMR: $\delta =$ 8.20–7.10 (m, 9H, C_6H_5, $C_6H_4NO_2$), 5.28 (s, H2), 5.12 (d, $J = 8.8$ Hz, H5), 3.05–2.94 (m, H4), 2.18 (s, NCH_3), 0.72–0.66 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, CHCH_3) ppm; ^{13}C NMR: $\delta = 151.3$ (CNO_2), 139.6, 133.5, 133.3, 130.3, 130.1, 128.6, 128.1, 126.6, 124.3 (C_{arom}), 92.2 (C2), 82.6 (C5), 63.6 (C4), 36.5 (N*C*H_3), 15.2 (CH*C*H_3) ppm.

(2R,4S,5R)-3,4-Dimethyl-2-(2-nitrophenyl)-5-phenyloxazolidine (**6b**, $C_{17}H_{18}N_2O_3$): ^{1}H NMR: $\delta = 8.20–7.10$ (m, 9H, C_6H_5, $C_6H_4NO_2$), 5.94 (s, H2), 5.32 (d, $J = 5.8$ Hz, H5), 3.60–3.51 (m, H4), 2.28 (s, NCH_3), 0.72–0.66 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, CHCH_3) ppm; ^{13}C NMR: $\delta = 150.8$ (CNO_2), 139.4, 135.6, 133.0, 129.8, 129.6, 128.3, 128.2, 126.6, 124.2 (C_{arom}), 90.7 (C2), 81.8 (C5), 61.0 (C4), 34.4 (NCH_3), 9.7 (CH*C*H_3) ppm.

2-(2-Chlorophenyl)-3,4-dimethyl-5-phenyloxazolidine (**7**)

(2S,4S,5R)-2-(2-Chlorophenyl)-3,4-dimethyl-5-phenyloxazolidine (**7a**, $C_{17}H_{18}ClNO$): ^{1}H NMR: $\delta =$ 7.12–7.90 (m, 9H, C_6H_5, C_6H_4Cl), 5.19 (s, H2), 5.10 (d, $J = 8.32$ Hz, H5), 2.92–2.98 (m, H4), 2.14 (s, NCH_3), 0.66–0.72 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, CHCH_3) ppm; ^{13}C NMR: $\delta = 139.5$ (C–Cl), 135.2, 129.9, 129.6, 129.4, 127.9, 127.8, 127.6, 127.1, 126.1 (C_{arom}), 93.8 (C2), 82.7 (C5), 63.7 (C4), 35.8(NCH_3), 15.0 (CH*C*H_3) ppm.

(2R,4S,5R)-2-(2-Chlorophenyl)-3,4-dimethyl-5-phenyloxazolidine (**7b**, $C_{17}H_{18}ClNO$): ^{1}H NMR: $\delta = 7.90–7.12$ (m, 9H, C_6H_5, C_6H_4Cl), 5.81 (s, H2), 5.47 (d, $J = 5.0$ Hz, H5), 3.63–3.55 (m, H4), 2.23 (s, NCH_3), 0.72–0.66 (m, 3H, overlapping hydrogens of (2*S*) and (2*R*) isomers, CHCH_3) ppm; ^{13}C NMR: $\delta = 139.0$ (C–Cl), 137.0, 134.0, 129.9, 129.4, 129.1, 128.0, 126.9, 125.8, 127.8 (C_{arom}), 90.9 (C2), 81.9 (C5), 61.9 (C4), 33.7 (N*C*H_3), 9.4 (CH*C*H_3) ppm.

Acknowledgement

Thanks are due to Dr. *G. Lautenschläger* and his team from Mikrowellen-Laborsysteme (MWS; Leutkirch/Germany) for special equipment and technical support and the Fonds der Chemischen Industrie for financial support.

References

[1] Gabriel C, Gabriel S, Grant EH, Halstead BSJ, Mingos DMP (1998) Chem Soc Rev **27**: 213
[2] Galema S (1997) Chem Soc Rev **26**: 233
[3] Kuhnert N (2002) Angew Chem **114**: 1943
[4] Strauss CR (2002) Angew Chem **114**: 3741
[5] Soliman S, Abdine H, El-Nenaey S (1975) Aust J Chem **28**: 49
[6] Beckett A, Jones G, Hollingsbee D (1978) J Pharm Pharmacol **30**: 15
[7] Bergmann E (1953) Chem Rev **53**: 309
[8] Pfanz H, Kirchner G (1958) Liebigs Ann Chem **614**: 149
[9] Neelankantan L (1971) J Org Chem **36**: 2256
[10] Neelankantan L, Molin-Case J (1971) J Org Chem **36**: 2261
[11] Just G, Potvin P, Uggowitzer P, Bird P (1983) J Org Chem **48**: 2923
[12] Berlan J, Besace Y, Pourcelot G, Cresson P (1983) J Organomet Chem **256**: 181
[13] Brunner H, Miehling W (1984) Monatsh Chemie **115**: 1237
[14] Real S, Kronenthal D, Wu H (1993) Tetrahedron Lett **34**: 8063
[15] Kuhnert N, Danks TN (2001) Green Chem **3**: 68
[16] Agami C, Rizk T (1985) Tetrahedron **41**: 537
[17] Beckett A, Jones G (1977) Tetrahedron **33**: 3313
[18] Soliman S (1973) Can J Pharm Sci **8**: 132