PREPARATION OF 5,6,7,8-TETRAHYDRO-7-ARYL-3-METHYLISOXAZOLO[4,5-*b*]AZEPIN-5(4*H*)-ONES

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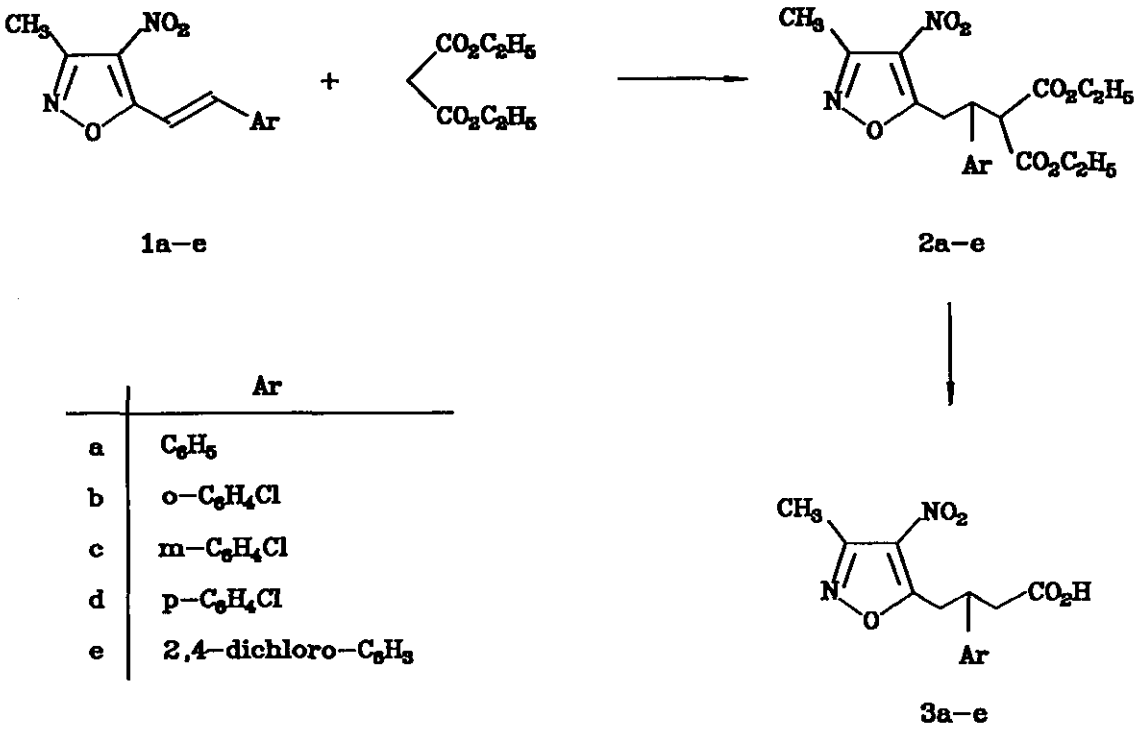
Abstract - 3-Methyl-4-nitro-5-styrylisoxazoles (**1a-e**) reacted with diethyl malonate to give the corresponding Michael adducts (**2a-e**) which have been hydrolyzed to 4-(3-methyl-4-nitroisoxazol-5-yl)-3-phenyl(chlorophenyl)butanoic acids (**3a-e**). Subsequent reduction of the nitro group of these acids afforded the title compounds (**4a-e**) by intramolecular cyclization.

A recent trend of organic chemistry is devoted to the study of heterocyclic compounds which could be considered as latent functional groups. Thus, isoxazolines[1] and oxazoles[2] were used for the preparation of natural products and carboxylic acids, respectively. In addition, isoxazoles were considered as masked ß-diketones,[3] α,ß-unsaturated ketones,[3] 1,4-diketones,[4] and ß-amino enones.[3] To this end we have already prepared several 3-methyl-4-nitro-5-styrylisoxazoles which gave cinnamic[5] and coumaric acids[6] via alkaline hydrolysis of the 3-methyl-4-nitroisoxazol-5-yl group which may be considered as a masked carboxylic function. Bromine addition to the double bond of the styryl portion led to 5-(1,2-dibromo-2-arylethyl)-3-methyl-4-nitroisoxazoles which in turn gave aryl propiolic acids.[7] Although the reactivity of 3-methyl-4-nitro-5-styrylisoxazoles as acceptors in the Michael reaction has already been described,[8-10] no isoxazolo[4,5-*b*]azepinones have been reported in the literature. Following our interest in this field, we wish now to describe the preparation of title compounds using 3-methyl-4-nitro-5-styrylisoxazoles (**1a-e**)[5] as starting materials.

RESULTS AND DISCUSSION

Heating of a mixture of compounds (**1a-e**) and diethyl malonate in presence of piperidine gave the corresponding Michael adducts (**2a-e**) as reported in Scheme 1.



	Ar
a	C_6H_5
b	o–C_6H_4Cl
c	m–C_6H_4Cl
d	p–C_6H_4Cl
e	2,4–dichloro–C_6H_3

It is worthy to note that when the molar ratio between (**1a**) and diethyl malonate was 1:3 the yield of (**2a**) was found to be 71% whereas higher yield (96%) was obtained when the above ratio was 1:10. The structures of compounds (**2a-e**) were determined by spectroscopic evidence; in particular, the presence of a triplet of doublets due to long range couplings ($^2J_{C5'\text{-}CH2}$ =6.6 Hz and $^3J_{C5'\text{-}H3}$ =3.4 Hz) for the carbon at position 5 of the isoxazole ring in the coupled ^{13}C-nmr spectra confirms the regiospecific attack of diethyl malonate on compounds (**1a-e**). In addition, the carbon atoms of the carbethoxy groups in compounds (**2a-e**) display six signals owing to the presence of a chiral center (Table 1). The same effect is reflected in the ^{1}H-nmr spectra; in fact, the two carbethoxy functions show two well separated signals both for the OCH_2 and CH_3 groups; in the mean time the 4-CH_2 protons appear as the AB part of an ABCD spin system which was analyzed for compounds (**2a-e**) by least-squares fitting thus obtaining the relative coupling constants and chemical shifts (Table 2).[11] The ^{13}C-nmr spectra of compounds bearing a chlorine atom at position 2 on the phenyl ring show a low frequency shift for C-3, C-2,

Table 1. ^{13}C-Nmr data of compounds **2a-e** and **3a-e** [a]

Compd	C=O	C-2	C-3	C-4	C-5'	C-4'	C-3'	OCH_2	CH_2CH_3	3'-CH_3	Others
2a	167.42 166.73	57.33	42.62	31.56	172.07	130.26	155.03	61.74 61.29	13.78 13.44	11.21	138.10(s), 128.37(d), 127.64(d), 127.52(d)
2b	167.31 166.67	55.75	38.82	30.20	171.72	130.39	155.17	61.89 61.55	13.82 13.50	11.33	135.76(s), 134.01(s), 129.94(d) ,128.79(d), 128.64(d),126.92(d)
2c	167.19 166.57	57.13	42.22	31.39	171.66	130.34	155.23	61.97 61.57	13.82 13.54	11.31	140.31(s) ,134.26(s) 129.78(d),128.00(d) 127.94(d),125.94(d)
2d	167.26 166.64	57.30	41.98	31.45	171.76	130.33	155.23	61.96 61.56	13.85 13.58	11.34	136.75(s), 133.56(s), 129.16(d), 128.70(d)
2e	167.10 166.54	55.62	38.34	30.03	171.41	130.41	155.28	62.00 61.72	13.83 13.57	11.35	134.78(s), 134.53(s), 134.01(s), 129.74(d), 129.54(d), 127.29(d)
3a	176.53	39.88	39.45	33.65	172.35	130.30	155.35	-	-	11.38	140.64(s), 128.79(d), 127.52(d), 126.88(d)
3b	176.34	38.41	35.65	31.95	171.89	130.46	155.38	-	-	11.41	137.85(s), 133.66(s), 130.10(d), 128.64(d), 127.52(d), 127.23(d)
3c	175.62	39.55	39.09	33.47	171.86	130.45	155.47	-	-	11.41	142.71(s), 134.64(s), 130.12(d), 127.83(d), 127.20(d), 125.15(d)
3d	176.42	39.86	38.81	33.50	171.95	130.39	155.45	-	-	11.41	139.06(s), 133.35(s), 129.00(d), 128.33(d)
3e	176.04	38.33	35.27	31.84	171.53	130.54	155.47	-	-	11.42	136.48(s), 134.41(s), 133.84(s), 129.92(d), 128.47(d), 127.58(d)

[a]Multiplicities determined by APT sequence and/or from coupled spectra.

and C-4 in **2b,e** and **3b,e**, and for C-7, C-6, and C-8 in compounds **4b,e** (Tables 1 and 3). This shift could be ascribed to both compressional and steric effects of the substituent; these latter are then emphasized in the ^{1}H-nmr spectra of **2b,e** by the appearance of diastereotopism in the OCH_2CH_3 groups at higher frequency which now appear as ABX_3 systems. In addition, in both compounds (**2b**) and (**2e**) dynamic effects are observed for H-2 and H-3.

The gas mass spectra of pure compounds (**2a-e**) show, beside the peaks attributable to the analyzed com-

Table 2. ^{1}H-Nmr data of compounds **2a-e** *($CDCl_3$, 300 MHz)*[a]

Compd	OCH_2	OCH_2'	H-3	H-2	H-4/ H-4'	3'-CH_3	CH_2CH_3	CH_2CH_3'	Aromatics
2a	4.257(q)	3.939(q)	4.080-4.012	3.842-3.586		2.440	1.307	0.983	7.350-7.150
			4.053	*3.827*	*3.797 3.622*				
2b[b]	4.333-4.182(m)	4.005(q)	4.672-4.588	3.995	3.860-3.726	2.463	1.296	1.038	7.311-7.118
			4.628	*3.995*	*3.801 3.773*				
2c	4.237(q)	3.972(q)	4.076-3.999	3.801-3.583		2.452	1.287	1.020	7.184-7.081
			4.020	*3.779*	*3.753 3.620*				
2d	4.250(q)	3.971(q)	4.080-3.990	3.816-3.572		2.458	1.299	1.034	7.233-7.119
			4.053	*3.782*	*3.780 3.616*				
2e[b]	4.333-4.180(m)	4.041(q)	4.628-4.542	3.940	3.850-3.711	2.483	1.297	1.091	7.332-7.324 7.195-7.189
			4.582	*3.940*	*3.794 3.760*				

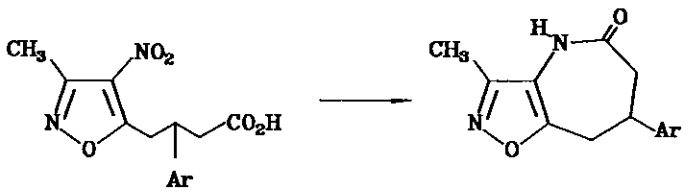
[a]Calculated spectra in italics. [b]Spectra recorded at 50°C.

pounds, other signals due to the corresponding styryl derivatives and to diethyl malonate; these results are in agreement with a retro-Michael reaction occurring in the injector. The expected 4-(3-methyl-4-nitroisoxazol-5-yl)-3-arylbutanoic acids (**3a-e**) were then obtained by refluxing compounds (**2a-e**) in diluted HCl. Finally, reduction of

Table 3. ^{13}C-Nmr data for compounds **4a-e**

Compd	C-3[a]	C-3a	C=O	C-6[b]	C-7[b]	C-8[b]	C-8a	Others
4a	153.44	116.07	173.35	43.75	35.87	34.19	156.19	143.43(s),128.89(d), 127.19(d),126.34(d), 9.17(q)
4b	153.60	116.21	173.38	42.43	32.46	32.74	156.01	140.67(s),133.04(s),129.89(d),128.40(d), 127.52(d),127.01(d),9.22(q)
4c	153.43	116.10	172.92	43.48	35.64	33.98	155.77	145.32(s),134.73(s),130.21(d),127.46(d), 126.68(d),124.64(d),9.20(q)
4d	153.11	115.98	172.54	43.53	35.27	33.93	155.90	141.66(s),133.12(s),129.08(d),127.76(d), 9.14(q),
4e	153.50	116.19	172.81	42.25	32.17	32.59	155.71	139.21(s),133.77(s),133.66(s),129.69(d), 127.90(d),127.82(d),9.18(q),

[a]Assigned on the basis of coupled spectra ($^{2}J_{C3\text{-}Me}$ = 7.0-7.2 Hz).
[b]Multiplicities determined by APT technique.



3a–e 4a–e

the nitro group of compounds (**3a-e**) with $SnCl_2.2H_2O$ and concentrated hydrochloric acid gave the title compounds (**4a-e**) through intramolecular ring closure between the formed amino group and the carboxylic function. The azepinone structures of compounds (**4a-e**) are unambiguosly confirmed by their spectroscopic data which are in agreement with those reported for similar compounds.[12] The distinction between C-3 and C-8a in com-

Table 4. ^{1}H-Nmr data of compounds **3a-e** and **4a-e**

Compd	*δ(300 MHz, $CDCl_3$)*
3a	7.316-7.127(m, 5H, Ph), 3.824-3.723(m, 1H, H-3), 3.678-3.493(m, 2H, H-4 and H-4'), 2.896-2.751 (m, 2H, H-2 and H-2'), and 2.485(s, 3H, 3'-CH_3)
3b	7.367-7.122(m, 4H, ArH_4), 4.393-4.293(m, 1H, H-3), 3.721-3.587(m, 2H, H-4 and H-4'), 2.948-2.800 (m, 2H, H-2 and H-2'), and 2.503(s, 3H, 3'-CH_3)
3c	7.330-7.107(m, 4H, ArH_4), 3.811-3.709(m, 1H, H-3), 3.655-3.476(m, 2H, H-4 and H-4'), 2.856-2.772 (m, 2H, H-2 and H-2'), and 2.513(s, 3H, 3'-CH_3)
3d	7.335-7.175(m, 4H, ArH_4), 3.822-3.721(m, 1H, H-3), 3.684-3.472(m, 2H, H-4 and H-4'), 2.802(d, J = 7.4 Hz, 2H, 2-CH_2), and 2.505(s, 3H, 3'-CH_3)
3e	7.380-7.114(m, 3H, ArH_3), 4.354-4.252(m, 1H, H-3), 3.708-3.572(m, 2H, H-4 and H-4'), 2.928-2.782 (m, 2H, H-2 and H-2'), and 2.520(s, 3H, 3'-CH_3)
4a	8.014(br s, exch., 1H, NH), 7.415-7.118(m, 5H, Ph), 3.490-3.382(m, 1H, H-7), 3.328-3.230(m, 1H), 3.208-3.085(m, 1H), 3.066-2.972(m, 1H), 2.968-2890(m, 1H), and 2.281(s, 3H, 3-CH_3)
4b	8.704(br s, exch., 1H, NH), 7.422-7.192(m, 4H, ArH_4), 3.970-3.862(m, 1H, H-7), 3.382-3.290(m, 1H), 3.126-3.010(m, 2H), 2.943-2.862(m, 1H), and 2.300(s, 3H, 3-CH_3)
4c	8.420(br s, exch., 1H, NH), 7.332-7083(m, 4H, ArH_4), 3.469-3.368(m, 1H, H-7), 3.328-3.235(m, 1H), 3.188-3.077(m, 1H), 3.048-2.960(m, 1H), 2.952-2.881(m, 1H), and 2.275(s, 3H, 3-CH_3)
4d	7.626(br s, exch., 1H, NH), 7.344-7145(m, 4H, ArH_4), 3.510-3.380(m, 1H, H-7), 3.352-3.213(m, 1H), 3.190-2.862(m, 3H), and 2.274(s, 3H, 3-CH_3)
4e	8.274(br s, exch., 1H, NH), 7.438-7126(m, 3H, ArH_3), 3.918-3.832(m, 1H, H-7), 3.368-3.280(m, 1H), 3.104-2.987(m, 2H), 2.893-2.841(m, 1H), and 2.296(s, 3H, 3-CH_3)

pounds (**4a-e**) is easily achieved from proton coupled ^{13}C-nmr spectra in which they appear as a quartet and a triplet of doublets, respectively.

In addition, compounds (**4a-e**) show a singlet near 116 ppm for the carbon at position 3a (Table 3) which, together with the NH signal in the ^{1}H-nmr spectrum (Table 4) and the stretching vibration of the amidic linkage at 1657-1665 cm^{-1}, are diagnostic for the assigned structures.

The presence of the isoxazole ring as latent functional group in **4a-e** may be also interesting to functionalize the azepinone ring.

EXPERIMENTAL

Melting points were determined on a Büchi 510 apparatus and are uncorrected. Infrared spectra were recorded with a Perkin-Elmer 283 instrument in potassium bromide pellets. ^{1}H- And ^{13}C-nmr spectra were taken in $CDCl_3$ on a Varian VXR-300 instrument; chemical shifts are reported in ppm high frequency from tetramethylsilane as secondary reference standard, coupling constants in Hz. Mass spectra were registered with an Hewlett-Packard 5790A instrument, whereas those of compounds (**4a-e**) were obtained at 70 eV by direct inlet (D.E.I.) of sample in a VG 70-70 EQ spectrometer. Analytical and preparative thin-layer chromatography (tlc) were carried out with Merck silica gel 60 F_{254} and mineral light lamp (model UV SL-58) was used for revealing spots.

General Procedure for the Preparation of Compounds **2a-e**

A mixture of the requisite compound (**1**) (50 mmol) and diethyl malonate (0.5 mol) was treated with piperidine (4 ml) and heated under stirring at 90 °C for 9 h. Removal of the solvent under reduced pressure (39-40 °C and 0.06 mmHg) left a sticky residue, to which was added methanol (5 ml) and set aside for 2-3 h. The solid obtained was filtered off, washed with the same solvent, and dried to give compound (**2**); a second crop of the same material was then obtained by concentration of the mother liquors which showed the presence (tlc analysis with benzene as eluant) of a very small amount of unreacted **1** (0.010-0.015 g) together with additional **2** and a third band at lower Rf which was no further investigated. Analytical samples were obtained by two recrystallizations from ethanol.

Ethyl 2-carbethoxy-4-(3-methyl-4-nitroisoxazol-5-yl)-3-phenylbutyrate **2a**: (96%); mp 72-73 °C. Ir ν_{max}: 1750, 1730, 1605, 1515, 1380, 1370, 826, 764, and 698 cm^{-1}; ms m/z (%): 390 (3.5) M^{+}, 345 (9.9), 299 (21.8), 265 (95), 160 (74.6), 131 (88), and 105 (100). Anal. Calcd for $C_{19}H_{22}N_2O_7$: C, 58.46; H, 5.68; N, 7.18. Found: C, 58.28; H, 5.68;

N, 7.37.

Ethyl 2-carbethoxy-3-(2-chlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butyrate **2b**: (87%); mp 56-57 °C. Ir ν_{max}: 1755, 1728, 1605, 1515, 1380, 1369, 820, and 766 cm^{-1}; ms m/z (%): 424 (0.1) M^+, 389 (20), 279 (44), 237 (100), and 165 (50). Anal. Calcd for $C_{19}H_{21}N_2O_7Cl$: C, 53.72; H, 4.98; N, 6.59. Found: C, 54.00; H, 5.00; N, 6.88.

Ethyl 2-carbethoxy-3-(3-chlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butyrate **2c**: (74%); mp 51-52 °C. Ir ν_{max}: 1748, 1723, 1607, 1517, 1381, 1368 and 828 cm^{-1}; ms m/z (%): 424 (0.1) M^+, 299 (18.4), 249 (10.1), 169 (29.9), 165 (91), 160 (100), 139 (85.9), 133 (32), and 115 (51.5). Anal. Calcd for $C_{19}H_{21}N_2O_7Cl$: C, 53.72; H, 4.98; N, 6.59. Found: C, 53.59; H, 5.04; N, 6.47.

Ethyl 2-carbethoxy-3-(4-chlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butyrate **2d**: (97%); mp 71-72 °C. Ir ν_{max}: 1750, 1722, 1610, 1522, 1383, 1370 and 822 cm^{-1}; ms m/z (%): 424 (0.1) M^+, 299 (46.9), 265 (24.9), 249 (19.3), 169 (37.9), 165 (87.9), 160 (34.4), 141 (40.5), 139 (100), 133 (13.9), and 115 (26.5). Anal. Calcd for $C_{19}H_{21}N_2O_7Cl$: C, 53.72; H, 4.98; N, 6.59. Found: C, 54.00; H, 4.97; N, 6.46.

Ethyl 2-carbethoxy-3-(2,4-dichlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butyrate **2e**: (70%); mp 54-55 °C. Ir ν_{max}: 1759, 1720, 1610, 1523, 1378, 1366, 827, and 766 cm^{-1}; ms m/z (%): 458 (0.2) M^+, 299 (20.6), 217 (21.6), 203 (44.2), 199 (53.2), 160 (100). Anal. Calcd for $C_{19}H_{20}N_2O_7Cl_2$: C, 49.69; H, 4.39; N, 6.10. Found: C, 49.47; H, 4.20; N, 6.26.

Hydrolysis of Compounds **2a-e**

Compound (**2**) (4 mmol) suspended in HCl (6 N, 16 ml) was heated under stirring at 125-130 °C. After 30 h the mixture was cooled, and the sticky solid separated was washed with water and then purified by dissolution in a saturated solution of $NaHCO_3$ (50 ml); re-precipitation with diluted HCl gave the acids (**3**). Analytical samples were obtained by recrystallization from diluted ethanol.

4-(3-Methyl-4-nitroisoxazol-5-yl)-3-phenylbutanoic Acid **3a**: (97%); mp 141-142 °C. Ir ν_{max}: 3300-2400br, 1702, 1603, 1525, 1380, 824, and 698 cm^{-1}. Anal. Calcd for $C_{14}H_{14}N_2O_5$: C, 57.93; H, 4.86; N, 9.65. Found: C, 57.85; H, 4.86; N, 9.72.

3-(2-Chlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butanoic Acid **3b**: (85%); mp 135-136 °C. Ir ν_{max}: 3300-2500br, 1700, 1601, 1517, 1379, 828, and 751 cm^{-1}. Anal. Calcd for $C_{14}H_{13}N_2O_5Cl$: C, 51.78; H, 4.04; N, 8.63.

Found: C, 51.62; H, 3.90; N, 8.82.

3-(3-Chlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butanoic Acid **3c**: (86%); mp 125-126 °C. Ir ν_{max}: 3500-2400br, 1700, 1599, 1518, 1380, and 826 cm^{-1}. Anal. Calcd for $C_{14}H_{13}N_2O_5Cl$: C, 51.78; H, 4.04; N, 8.63. Found: C, 51.63; H, 3.99; N, 8.81.

3-(4-Chlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butanoic Acid **3d**: (85%); mp 129-130 °C. Ir ν_{max}: 3500-2300br, 1700, 1605, 1530, 1385, and 826 cm^{-1}. Anal. Calcd for $C_{14}H_{13}N_2O_5Cl$: C, 51.78; H, 4.04; N, 8.63. Found: C, 51.94; H, 3.94; N, 8.63.

3-(2,4-Dichlorophenyl)-4-(3-methyl-4-nitroisoxazol-5-yl)butanoic Acid **3e**: (82%); mp 152-153 °C. Ir ν_{max}: 3300-2400br, 1696, 1598, 1514, 1375, 827, 793, and 764 cm^{-1}. Anal. Calcd for $C_{14}H_{12}N_2O_5Cl_2$: C, 46.82; H, 3.37; N, 7.80. Found: C, 46.55; H, 3.28; N, 7.93.

General Procedure for the Preparation of Compounds **4a-e**

To compound (**3**) (3 mmol) in ethanol (10 ml) was added concentrated HCl (3 ml), $SnCl_2.2H_2O$ (2.1 g; 9 mmol), and Sn (1.1 g; 9 mmol); the mixture was then refluxed under stirring for 1 h. Removal of the solvent left a sticky residue, to which was added H_2O (25 ml) and extracted with ether (3 x 60 ml). The ethereal extracts were neutralized with a saturated solution of $NaHCO_3$, washed with H_2O, and dried over Na_2SO_4; evaporation to dryness gave a gummy product which was worked up with EtOH to afford a small amount (0.05-0.15 g) of compound (**4**) as a colorless solid. Preparative chromatography of the mother liquors ($CHCl_3$-MeOH 10:1 v/v, as eluant) gave additional **4**. Analytical samples were obtained by recrystallization from ethanol.

5,6,7,8-Tetrahydro-3-methyl-7-phenylisoxazolo[4,5-*b*]azepin-5(4*H*)-one **4a**: (40%); the solid darkened near 236 °C and melted at 239-240 °C (decomp). Ir ν_{max}: 3320-2650, 1665, 1608, and 703 cm^{-1}; ms m/z (%): 242 (46.2) M^+, 131 (38), 111 (72.4), and 104 (100). Anal. Calcd for $C_{14}H_{14}N_2O_2$: C, 69.41; H, 5.82; N, 11.56. Found: C, 69.13; H, 5.71; N, 11.80.

5,6,7,8-Tetrahydro-7-(2-chlorophenyl)-3-methylisoxazolo[4,5-*b*]azepin-5(4*H*)-one **4b**: (35%); mp 197-198 °C. Ir ν_{max}: 3340-2800, 1665, and 749 cm^{-1}; ms m/z (%): 276 (36.5) M^+, 241 (13.6), 165 (23.1), 138 (81), 111 (100), and 103 (27.4). Anal. Calcd for $C_{14}H_{13}N_2O_2Cl$: C, 60.77; H, 4.74; N, 10.12. Found: C, 61.01; H, 4.49; N, 10.14.

5,6,7,8-Tetrahydro-7-(3-chlorophenyl)-3-methylisoxazolo[4,5-*b*]azepin-5(4*H*)-one **4c**: (34%); mp 182-183 °C. Ir

ν_{max}: 3340-2750, 1657, and 1596 cm^{-1}; ms m/z (%): 276 (43.8) M^+, 165 (21.9), 138 (75), 111 (100), and 103 (22). Anal. Calcd for $C_{14}H_{13}N_2O_2Cl$: C, 60.77; H, 4.74; N, 10.12. Found: C, 60.49; H, 4.76; N, 9.85.

5,6,7,8-Tetrahydro-7-(4-chlorophenyl)-3-methylisoxazolo[4,5-*b*]azepin-5(4*H*)-one **4d**: (30%); the solid darkened near 262 °C and melted at 271-272 °C (decomp). Ir ν_{max}: 3360-2750, 1665, and 829 cm^{-1}; ms m/z (%): 276 (40) M^+, 165 (28.8), 138 (100), 111 (91.5), and 103 (22). Anal. Calcd for $C_{14}H_{13}N_2O_2Cl$: C, 60.77; H, 4.74; N, 10.12. Found: C, 60.56; H, 4.71; N, 10.15.

5,6,7,8-Tetrahydro-7-(2,4-dichlorophenyl)-3-methylisoxazolo[4,5-*b*]azepin-5(4*H*)-one **4e**: (34%); the sample darkened near 212 °C and melted at 218-220 °C (decomp). Ir ν_{max}: 3360-2750, 1664, 799, and 827 cm^{-1}; ms m/z (%): 310 (31) M^+, 199 (16), 172 (63.4), 137 (16.5), and 111 (100). Anal. Calcd for $C_{14}H_{12}N_2O_2Cl_2$: C, 54.04; H, 3.89; N, 9.00. Found: C, 53.75; H, 3.84; N, 9.28.

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