Asian Journal of Chemistry Vol. 11, No. 2 (1999), 441–449

Synthesis and Spectroscopic Properties of 2-[1-Benzyl-1,2,3-Triazolo-4]-5-Aryl-1,3,4-Oxadiazoles

HASAN TASHTUOSH, SULTAN ABU-ORABI,
EMAN TA'AN and MAHMOUD AL-TALIB*
Department of Chemistry, Yarmouk University, Irbid, Jordan

A series of 2-[1-benzyl-1,2,3-triazolo-4]-5-aryl-1,3,4-oxadiazoles **5a–l** were prepared *via* phosphorus oxychloride promoted dehydration of the corresponding carbohydrazides, **4a–l**. The products were fully identified by NMR, IR, UV and MS spectroscopy and elemental analysis.

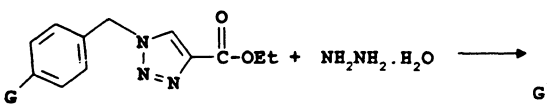
INTRODUCTION

Substituted 1,3,4-oxadiazoles attracted considerable attention in organic synthesis due to their broad range of biological, medicinal, agricultural and industrial applications[1–5]. The subject has recently been reviewed[6]. Despite the intensive research on substituted mono-, bis-, and tris-1,3,4-oxadiazoles[7–10], little is known about 1,3,4-oxadiazoles attached to heterocyclic rings with more than one heteroatom. That prompted us to study the synthesis and characterization of a series of compounds which possess a 1,2,3-triazole moiety linked to an oxadiazole ring.

RESULTS AND DISCUSSION

The starting materials, 1-substituted benzyl-4-hydrazinocarbonyl-1,2,3-triazoles **2a–d** were prepared by the reaction of ethyl substituted benzyl-1H-1,2,3-triazole-4-carboxylate (**1a–d**) with hydrazine monohydrate (Scheme-1). The latter compounds were characterized by IR, ^{1}H NMR and elemental analyses. Physical and spectroscopic properties of compounds **2a–d** are given in Table-1.

Scheme-1



1 a-d 2 a-d

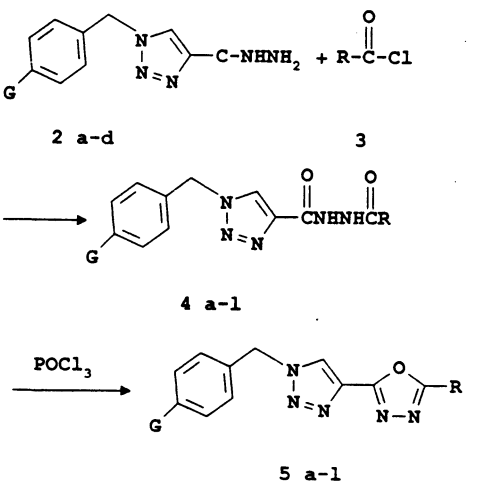
1, 2	G
a	H
b	CH_3
c	OCH_3
d	Cl

TABLE-1
MELTING POINTS, YIELDS, IR AND ^{1}H—NMR SPECTROSCOPIC DATA FOR COMPOUNDS 2a–d

Comp. No.	m.p. (°C)	Yield (%)	IR (cm^{-1}) KBr disc	^{1}H NMR, (d_6-DMSO/TMS), δ (ppm), J (Hz)
2a	184–5	85	3290, 1668, 1450	4.46 (bs, 2H); 5.65 (s, 2H); 7.36 (s, 5H); 8.60 (s, H); 9.69 (bs, 1H)
b	177–8	92	3260, 1670, 1440	2.29 (s, 3H); 4.44 (bs, 2H); 5.59 (s, 2H); 7.22 (s, 4H); 8.56 (s, 1H); 9.67 (bs, H)
c	180–1	78	3260, 1670, 1445	3.74 (s, 3H); 4.45 (bs, 2H); 5.56 (s, 2H); 6.93 (d, J = 8.7 Hz, 2H); 7.33 (d, J = 8.7 Hz); 8.55 (s, 1H); 9.68 (bs, H)
d	186–7	91	3240, 1670, 1440	4.47 (bs, 2H); 5.66 (s, 2H); 7.42 (s, 4H); 8.63 (s, 1H); 9.70 (bs, 1H)
e	165–6	89	3250, 1660, 1440	4.47 (bs, 2H); 5.77 (s, 2H); 7.49–7.32 (m, 4H); 8.56 (s, 1H); 9.70 (bs, 1H)

Compounds **2a–d** readily reacted with one equivalent of acyl chlorides **3** in THF/water mixture and in the presence of potassium carbonate to afford 1-substituted benzyl-1,2,3-triazolo-4-[2-aroyl] carbohydrazides **4a–l** in good yields, Scheme-2.

Scheme-2



4, 5	G	R
a	H	C_6H_5
b	H	*p*-$CH_3OC_6H_4$
c	H	*p*-$CH_3C_6H_4$
d	Cl	*p*-$CH_3C_6H_4$
e	Cl	2-furyl
f	Cl	2-thienyl
g	CH_3	C_6H_5
h	CH_3	*p*-$CH_3OC_6H_4$
i	CH_3	*p*-$CH_3C_6H_4$
j	CH_3	2-furyl
k	CH_3	2-thienyl
l	CH_3O	C_6H_5

Table-2 includes names, molecular formulae and elemental analysis data of compounds **4a–l**, whereas Table-3 includes melting points, yields, IR, ^{1}H NMR and some ^{13}C NMR data of compounds **4a–l**. In IR spectra compounds **4a–l** exhibited the characteristic absorptions in the ranges 3320–3200 cm^{-1} assigned to the N—H stretching frequency, 1690–1630 cm^{-1} for the carbonyl groups.

TABLE-2
NAMES, MOLECULAR FORMULAE AND ELEMENTAL ANALYSES FOR COMPOUNDS **4a–l**

Comp. No.	Name	Mol. formula (m.w. g/mol)	Elemental analysis (%) Calcd. (found)		
			C	H	N
4a	1-Benzyl-1,2,3-triazolo-4- (2^1-benzoyl)carbohydrazide	$C_{17}H_{15}N_5O_2$ (321.3)	63.54 (63.69)	4.71 (4.83)	21.79 (21.60)
b	1-Benzyl-1,2,3-triazolo-4- [2^1-(*p*-anisoyl)] carbohydrazide	$C_{18}H_{17}N_5O_3$ (351.4)	61.53 (61.38)	4.88 (4.95)	19.93 (19.81)
c	1-Benzyl-1,2,3-triazolo-4- [2^1-(*p*-toluoyl)] carbohydrizide	$C_{18}H_{17}N_5O_2$ (335.4)	64.47 (64.21)	5.11 (5.33)	20.88 (21.08)
d	1-Benzyl-1,2,3-triazolo-4- (2^1-benzoyl) carbohydrizide	$C_{15}H_{13}N_5O_2$ (327.4)	55.04 (55.07)	4.00 (4.22)	21.39 (21.42)
e	1-(*p*-chlorobenzyl)-1,2,3-triazo lo-4-[2′-(*p*-anisoyl)] carbohydrazide	$C_{18}H_{16}ClN_5O_3$ (385.8)	56.04 (55.90)	4.18 (4.40)	18.15 (18.06)
f	1-(*p*-chlorobenzyl)-1,2,3-triazo lo-4-[2′-(*p*-furyl carbonyl)] carbohydrazide	$C_{15}H_{12}ClN_5O_2$ (345.8)	52.11 (52.34)	3.49 (3.69)	20.26 (20.35)
g	1-(*p*-methylbenzyl)-1,2,3-triazo lo-4-(2′-benzoyl) carbohydrazide	$C_{18}H_{17}N_2O_2$ (335.4)	64.47 (64.20)	5.11 (5.09)	20.88 (20.86)
h	1-(*p*-methylbenzyl)-1,2,3-triazo lo-4-[2′-(*p*-anisoyl)] carbohydrazide	$C_{19}H_{19}N_5O_3$ (365.4)	62.46 (62.34)	5.24 (5.05)	19.17 (19.30)
i	1(*p*-methylbenzyl)-1,2,3-triazo lo-4-[2′-(*p*-toluoyl)] carbohydrazide	$C_{19}H_{19}N_5O_2$ (349.4)	65.32 (65.51)	5.48 (5.19)	20.44 (20.39)
j	1-(*p*-methylbenzyl)-1,2,3-triazo lo-4-[2′-furylcabon-yl)] carbohydrazide	$C_{16}H_{15}N_5O_3$ (325.3)	59.07 (58.87)	4.65 (4.90)	21.53 (21.49)
k	1-(*p*-methylbenzyl)-1,2,3-triazo lo-4-[2′-(2-thienylcarbonyl)] carbohydrazide	$C_{16}H_{15}N_5O_2S$ (341.8)	56.29 (56.32)	4.42 (4.34)	20.52 (20.54)
l	1-(*p*-methoxybenzyl)-1,2, 3-triazolo-4-[2′-benzoyl)] carbohydrazide	$C_{18}H_{17}N_5O_3$ (351.4)	61.53 (61.45)	4.88 (4.94)	19.93 (19.10)

In addition, ^{1}H NMR spectroscopy confirmed the structures of these compounds. Thus, the NH protons appeared as broad signal in the range δ 10.30–10.67. These protons underwent rapid hydrogen-deuterium exchange. The proton of the triazole ring appeared as a singlet in the range δ 8.68–8.79, whereas the benzylic protons attached to the triazole ring resonate at δ 5.61–5.72. Other protons resonate at the expected shift values (Table-3).

TABLE-3
m.p. YIELD, IR, ^{1}H NMR AND ^{13}C NMR DATA FOR COMPOUNDS **4a–l**

Comp. No.	m.p. (°C)	Yield (%)	IR (cm^{-1}) KBr disc	^{1}H NMR, (d_6-DMSO/TMS), δ (ppm), J (Hz)	^{13}C NMR (d^6-DMSO/TMS) δ (ppm), J (Hz)
4a	241–2	81	3250, 1670	5.70 (s, 2H) 7.32 (s, 5H); 7.96–7.38 (m, 5H); 8.77 (s, 1H); 10.50 (bs, 2H)	53.1, 127.1, 127.4, 128.0, 128.2, 128.4, 128.8, 131.8, 132.3, 135.5, 141.3, 159.1, 165.5
b	210–1	78	3300, 1690	3.83 (s, 3H); 5.69 (s, 2H); 7.04 (d, J = 8.7 Hz, 2H); 7.38 (s, 5H); 7.91 (d, J = 8.7 Hz, 2H); 8.76 (s, 1H); 10.39 (bs, 2H)	
c	220–2	82	3280, 1670	2.38 (s, 3H); 5.70 (s, 2H); 7.26–7.38 (b, 7H); 7.82 (d, J = 8.3 Hz, 2H); 8.76 (s, 1H); 10.43 (bs, 2H)	
d	262–3	85	3240, 1660	10.49 (bs, 2H); 8.76 (s, 1H); 7.84 (d, J = 4.8 Hz, 2H); 7.38 (s, 5H); 7.19 (t, J = 4.6 Hz, 1H); 5.69 (s, 2H)	
e	230–1	79	3320, 1650	3.83 (s, 3H); 5.67 (s, 2H); 7.04 (d, J = 8.6 Hz, 2H); 7.44 (s, 4H); 7.92 (d, J = 8.6 Hz, 2H); 8.79 (s, 1H); 10.42 (bs, 2H)	
f	237–8	86	3240, 1670	5.70 (s, 2H); 6.67 (b, 1H); 7.26 (d, J = 3.3 Hz, 1H); 7.44 (s, 4H); 7.91 (b, 1H); 8.78 (s, 1H); 10.43 (bs, 2H)	159.0, 157.1, 146.2, 145.6, 141.3, 134.4, 133.0, 129.9, 128.8, 127.2, 114.5, 111.8, 52.3
g	228–9	80	3260, 1660	2.29 (s, 3H); 5.70 (s, 2H); 8.74 (s, 1H); 8.00–7.24 (m, 9H); 10.49 (bs, 2H)	
h	196–7	86	3320, 1690	2.29 (s, 3H); 3.83 (s, 3H); 5.63 (s, 2H); 7.04 (d, J = 8.7 Hz, 2H); 7.24 (s, 4H); 7.89 (d, J = 8.7 Hz, 2H); 8.71 (s, 1H); 10.36 (bs, 2H)	
i	232–3	89	3245, 1660	2.37 (s, 6H); 5.64 (s, 2H); 7.30 (d, J = 8.6 Hz, 2H); 7.83 (d, J = 8.6 Hz, 2H); 8.70 (s, 1H); 10.39 (bs, 2H)	20.6, 20.9, 52.9, 126.9, 127.4, 128.0, 128.9, 129.3, 129.6, 132.5, 137.6, 141.3, 141.7, 159.1, 165.4
j	182–3	82	3260, 1660	2.29 (s, 3H); 5.62 (s, 2H); 6.66 (b, 1H); 7.24 (s, 5H); 7.91 (b, 1H); 8.69 (s, 1H); 10.41 (bs, 2H)	
k	244–5	94	3260, 1660	2.29 (s, 3H); 5.62 (s, 2H); 7.18 (d, J = 2.0 Hz, 2H); 7.29–7.21 (m, 3H); 7.82 (d, J = 4.9 Hz, 1H); 7.87 (d, J = 3.5 Hz, 1H); 8.69 (s, 1H); 10.44 (bs, 2H)	
l	225–6	87	3260, 1670	3.75 (s, 3H); 5.61 (s, 2H); 6.95 (d, J = 8.7 Hz, 2H); 7.35 (d, J = 8.7 Hz, 2H); 7.69–7.42 (m, 5H); 8.68 (s, 1H); 10.41 (bs, 2H)	

Treatment of compounds **4a–l** with excess phosphorus oxychloride in acetonitrile yielded the corresponding substituted 1,3,4-oxadiazoles, **5a–l**, scheme-2. The reactions proceeded smoothly to give very good yields (65–85%) of compounds **5a–l**. The structures of compounds **5a–l** were confirmed on the basis of their IR, ^{1}H NMR, ^{13}C NMR, UV and mass spectral data in addition to elemental analysis (Tables-4, 5 and 6). In IR spectra compounds **5a–l** show the typical absorptions of the oxadiazole ring at 1630–1590 cm^{-1} and 1030–1020 cm^{-1} (C=N and C—O respectively), which are recorded with medium intensity[6].

Compounds **5** show in ^{1}H NMR spectra peaks for the aromatic protons at the expected shift values. The benzylic protons attached to the ring resonate at δ 5.69–5.78, whereas the proton on the triazole ring resonated at lower field (δ 9.05–9.15). The chemical shift of this proton is found to be strongly solvent dependent, thus, in $CDCl_3$ it resonated at higher field (δ 7.50–8.20).

In ^{13}C NMR the oxadiazole ring exhibits characteristic signals for C-2 and C-5; the chemical shift of C-2 appeared around 157 δ, whereas the chemical shifts of C-5 showed strong dependence on the nature of the substituent. It resonates in the range δ 160–163, depending on the electronic effect of the substituent. ^{13}C NMR data is given in Table-5.

The UV spectra of compounds **5a–l** consist mainly of one absorption band at λ_{max}255–284 nm. This band is characteristic of 2-aryl-1,3,4-oxadiazoles[11].

In the mass spectra of compounds **5a–l**, the fragmentation pattern is in full agreement with that expected for 1,3,4-oxadiazole and 1,2,3-triazole ring systems[12, 13]. Thus these compounds give peaks corresponding to $[M^+-N_2]$ and $[M^+$-ArCNO], besides the molecular ion peaks $[M^+]$. In addition, the fragments $[ArCN]^+$ $[ArCNO]^+$, $[ArCO]^+$ and $PhCH^{2+}$ were observed. The mass spectral data is given in Table-6.

EXPERIMENTAL

Ethyl substituted benzyl-1H-1,2,3-triazole-4-carboxylates, **1a–d**, were prepared following a literature procedure[14, 15].

Melting points were determined on an electrothermal-digital melting point apparatus and were uncorrected. Infrared spectra were recorded on a PY-Unicam SP 300 spectrophotometer as KBr discs. ^{1}H NMR spectra were recorded on a Brucker WP 80-SY, WM-250 and AC-250 spectrometers in deuterated chloroform or dimethyl sulfoxide with tetramethyl silane (TMS) as an internal standard. ^{13}C NMR spectra were recorded on Brucker WM-250 and AC-250 spectrometers. Ultraviolet spectra were recorded in acetonitrile using a computerized CARY 2390 spectrophotometer linked to DS-15 data station. Mass spectra were recorded on a sector field double focusing unit VG 7070E mass spectrometer interfaced with PDP-11 mini computer for control and data acquisition.

Elemental analyses were performed at MHW Laboratories, Phoenix, Arizona, USA and CHN-Mikroanalysis Laboratory, Fakultat fur Chemie Universitat Konstanz, Germany. Chemicals were purchased from Aldrich or Fluka and were used without further purification.

TABLE-4
NAMES, MOLECULAR FORMULAE AND ELEMENTAL ANALYSIS OF COMPOUNDS 5a–l

Comp. No.	Name	Mol. formula (m. w.)	Elemental analysis (%) Calcd. (found)		
			C	H	N
5a	2-[1-Benzyl-1,2,3-triazolo-4]-5-phenyl-1,3,4-oxadiazole	$C_{17}H_{13}H_5O$ (303.3)	67.32 (67.36)	4.32 (4.55)	23.09 (22.85)
b	2-[1-Benzyl-1,2,3-triazolo-4]-5-(*p*-anisyl)-1,3,4-oxdi-azole	$C_{18}H_{15}N_5O_2$ (333.4)	64.86 (64.67)	4.54 (4.42)	21.01 (21.11)
c	2-[1-Benzyl-1,2,3-triazolo-4]-5-(*p*-tolyl)-1,3,4-oxadiazole	$C_{18}H_{15}NSO$ (317.4)	68.13 (67.80)	4.76 (5.00)	22.07 (21.97)
d	2-[1-(*p*-benzyl-1,2,3-triazolo-4]-5-(2-thienyl)-1,3,4-oxadiazole	$C_{15}H_{11}N_5OS$ (309.4)	58.24 (58.14)	3.58 (3.40)	22.64 (22.66)
e	2-[1-(*p*-chlorobenzyl)-1,2,3-triazolo-4]-5-(*p*-anisyl)-1,3,4-oxadiazole	$C_{18}H_{14}ClN_5O_2$ (367.8)	58.78 (58.59)	3.84 (4.04)	19.04 (19.16)
f	2-[1-(*p*-chlorobenzyl)-1,2,3-triazolo-4]-5-(2-furyl)-1,3,4-oxadiazole	$C_{15}H_{10}ClN_5O_2$ (327.7)	54.97 (54.63)	3.08 (3.40)	21.37 (20.94)
g	2-[1-(*p*-methylbenzyl)-1,2,3-triazolo-4]-5-phenyl)-1,3, 4-oxadiazole	$C_{18}H_{15}N_5O$ (317.4)	68.13 (67.91)	4.76 (4.56)	22.07 (22.16)
h	2-[1-(*p*-methylbenzyl)-1,2,3-triazolo-4]-5-(*p*-anisyl)-1,3,4-oxadiazole	$C_{19}H_{17}N_5O_2$ (347.4)	65.69 (65.49)	4.93 (4.75)	20.16 (20.13)
i	2-[1-(*p*-methylbenzyl)-1,2,3-triazolo-4]-5-(*p*-tolyl)-1,3,4-oxadiazole	$C_{19}H_{17}N_5O$ (331.4)	68.87 (68.76)	5.17 (5.40)	21.13 (20.82)
j	2-[1-(*p*-methylbenzyl)-1,2,3-triazolo-4]-5-(2-furyl)-1,3,4-oxadiazole	$C_{16}H_{13}N_5O_2$ (307.3)	62.53 (62.69)	4.26 (4.34)	22.78 (22.53)
k	2-[-*p*-methylbenzyl)-1,2,3-triazolo-4]-5-(2-thienyl)-1,3,4-oxadiazole	$C_{16}H_{13}N_5OS$ (323.8)	59.35 (59.36)	4.05 (4.13)	21.63 (21.95)
l	2-[1-(*p*-methoxybenzyl)-1,2,3-triazolo-4]-5-phenyl-1,3,4-oxadiazole	$C_{18}H_{15}N_5O_2$ (333.4)	64.86 (64.85)	4.54 (4.63)	21.01 (20.88)

TABLE-5

m.p. YIELD, IR, ^{1}H NMR AND ^{13}C NMR DATA OF COMPOUNDS **5a–l**

Comp No.	m.p. (°C)	Yield (%)	IR (cm^{-1}) KBr disc	^{1}H NMR, (d_6-DMSO/TMS) δ (ppm), J (Hz)	^{13}C NMR (d_6-DMSO/TMS) δ (ppm), J (Hz)
5a	155–6	85	1625, 1550, 1020	5.71 (s, 2H), 8.71–7.25 (m, 10H); 9.12 (s, 1H)	
b	180–1	90	1635, 1617, 1020	3.88 (s, 3H), 5.77 (s, 2H); 9.11 (s, 1H); 7.18 (d, J = 8.8 Hz, 2H); 7.41 (s, 5H); 8.08 (d, J = 8.8 Hz, 2H)	163.7, 162.2,157.5, 135.3, 132.9, 128.5, 128.3, 128.1, 126.3, 115.4, 114.9, 55.5, 53.4
c	185–6	88	1620, 1020	2.41 (s, 3H); 5.77 (s, 2H); 7.44 (b, 6H), 7.97 (d, J = 7.8 Hz, 2H); 9.08 (s, 1H)	21.2, 54.5, 123.4, 124.4, 127.2, 128.5, 129.1, 130.1, 131.4, 132.0, 134.2, 139.4, 158.1, 164.7
d	171–2	88	1630, 1590, 1020	5.76 (s, 2H); 7.33–7.28 (m, 1H); 7.41 (s, 5H); 9.14 (s, 1H); 7.93–7.88 (m, 2H)	54.7, 124.6, 128.3, 128.4, 129.3, 129.4, 130.4, 130.6, 133.4, 134.0, 141.4, 157.5 161.0
e	202–3	88	1620, 1600, 1030	3.88 (s, 3H); 5.78 (s, 2H); 7.18 (d, J = 8.6 Hz, 2H); 7.47 (s, 4H); 8.03 (d, J = 8.6 Hz, 2H); 9.13 (s, 1H)	53.9, 55.5, 114.5, 115.8, 124.3, 129.0, 129.6, 129.7, 132.1, 134.6, 135.4, 157.5,1 62.6, 164.7
f	184–5	84	1658, 1620, 1020	5.77 (s, 2H); 6.86 (m, 2H); 7.46 (s, 4H); 8.11 (b, 1H); 9.15 (s, 1H)	159.2, 156.6, 147.1, 138.3, 134.2, 133.4, 133.1, 132.5, 130.1, 126.6, 114.8, 112.7, 52.5
g	169–70	86	1655, 1615, 1020	2.28 (s, 3H); 5.70 (s, 2H); 7.21 (d, J = 7.9 Hz, H); 7.32 (d, J = 7.9 Hz, 2H); 7.64 (b, 3H); 8.08 (b, 2H); 9.09 (s, 1H)	163.6, 157.9, 137.7, 132.7, 132.3, 132.1, 129.4, 129.3, 128.1, 126.6, 126.6, 126.3,1 23.1
h	172–3	80	1630, 1610, 1040	2.32 (s, 3H); 3.88 (s, 3H); 5.70 (s, 1H); 7.25 (d, J = 8.7 Hz, 2H); 8.02 (d, J = 8.7 Hz, 2H); 8.20 (s, 4H); 9.09 (s, 1H)	
i	172–3	87	1650, 1590, 1020	2.30 (s, 3H); 2.41 (s, 3H); 5.70 (s, 2H); 7.29 (s, 4H); 7.44 (d, J = 8.5 Hz, 2H); 7.98 (d, J = 8.5 Hz, 2H); 9.10 (s, 1H)	163.8, 157.7, 142.3, 137.8, 132.9, 132.3, 129.9, 129.3, 128.1, 126.6, 126.2, 120.4, 53.2, 21.1, 20.6
j	148–9	94	1615, 1530, 1030	2.29 (s, 3H); 5.69 (s, 2H); 6.83 (b, 1H); 7.45 (m, 5H); 8.11 (b, 1H); 9.10 (s, 1H)	

Comp No.	m.p. (°C)	Yield (%)	IR (cm^{-1}) KBr disc	^1H NMR, (d_6-DMSO/TMS) δ (ppm), J (Hz)	^{13}C NMR (d_6-DMSO/TMS) δ (ppm), J (Hz)
k	161–2	89	1630, 1590, 1040	2.29 (s, 3H); 5.69 (s, 2H); 7.88–7.19 (m, 5H); 7.90 (b, 1H); 7.99 (b, 1H); 9.09 (s, 1H)	21.2, 54.5, 124.5, 124.7, 128.3, 128.5, 130.1, 130.4, 130.6, 133.9, 139.4, 157.5,1 60.9
l	163–4	89	1635, 1615, 1030	3.76 (s, 3H) 5.69 (s, 2H); 6.96 (d, J = 8.6 Hz, 2H); 8.12–7.66 (m, 5H); 9.08 (s, 1H)	

TABLE-6
UV AND MASS SPECTRAL DATA OF COMPOUNDS **5a–l**

Compound No.	UV (λ_{max}, nm)	MS (m/z)
5a	264, 194	303(M^+), 275, 198, 119, 105, 103, 91, 77
b	279	333(M^+), 305, 135, 133, 91
c	269	317(M^+), 289, 119, 117, 91, 77
d	284	309(M^+), 281, 109, 111, 91, 83
e	279	367(M^+), 339, 149, 133, 111, 90
f	276	327(M^+), 299, 125, 111, 95
g	255	317(M^+), 289, 119, 105, 103, 91, 77
h	279	347(M^+), 219, 133, 105, 90
i	268	331(M^+), 303, 198, 149, 133, 117, 105, 91
j	275	307(M^+), 198, 105, 93, 91, 77
k	280	323(M^+), 295, 198, 109, 105, 91
l	255	333(M^+), 305, 121, 119, 107, 105, 103, 91

General procedure for the preparation of 1-substituted benzyl-4-hydrazinocarbonyl-1,2,3-triazole, 2a–d

Hydrazine monohydrate (0.75 g, 15 mmol) was added dropwise, with stirring, to a boiled solution of ethyl substituted benzyl-1H-1,2,3-triazole-4-carboxylate, **1a–d** (5 mmol) in ethanol (50 mL). The resulting mixture was heated under reflux for 1 h. The solvent was removed under reduced pressure and the resulting solid was recrystallized from ethanol-tetrahydrofuran to give products **2a–d** as colorless needles. Table-1 presents melting points, yields, IR data and ^1H NMR for compounds **2a–d**.

General procedure for the preparation of 1-substituted benzyl-1,2,3-triazolo-4-[2′-aroyl]-carbohydrazides, 4a–l

A solution of acyl chloride (3) (10 mmol) in tetrahydrofuran (20 mL) was

added dropwise to a stirred solution of substituted benzyl-4-hydrazino carbonyl-1,2,3-triazole, **2a–e** (10 mmol) and potassium carbonate (10 mmol) in tetrahydrorufan (40 mL) and water (120 mL). The reaction mixture was stirred at room temperature for several hours. The resulting solid was collected by filteration and washed with water. Recrystallization from dimethylsulfoxide-water (1 : 3) gave compounds **4a–l** as colorless powders.

Table-2 includes names, molecular formulas and elemental analysis data of compounds **4a–l**. Melting points, yields, IR, ^{1}H NMR and some ^{13}C NMR data of these compounds are given in Table-3.

General procedure for the preparation of substituted 1,3,4-oxadiazoles, 5a–l

A solution of substituted carbohydrazides, **4a–l** (7.5 mmol) in acetonitrile (40 mL) and phosphorus oxychloride (20 g, 133 mmol) was heated under reflux for 3–5 h.

The reaction mixture was cooled to room temperature and poured into 200 mL of icewater slush. The resulting solid was collected by filtration, washed with water and recrystallized from chloroform-hexane (2 : 3).

Table-4 summarizes names, yields, molecular formulas and elemental analysis data of compounds **5a–l**. Melting points, yields, IR, ^{1}H NMR and ^{13}C NMR data of the above compounds are given in table-5, whereas UV and mass spectral data are given in Table-6.

ACKNOWLEDGEMENTS

We would like to thank Alexander von Humboldt Foundation (Germany) for instruments donation and Yarmouk University for financial support.

REFERENCES

1. F. Pochat, *Synthesis,* 146 (1984).
2. B.N. Goswani, J.C.S. Kataky and J.N. Barouah, *J. Heterocyclic Chem.*, **21,** 1225 (1984).
3. S.P. Summan and S.C. Bahel, *Agric. Biol. Chem.*, **43,** 1339 (1979).
4. S.B. Valenti, Pat. 1550440 (1979); *Chem. Abstr.*, **92,** 199758 (1980).
5. A. Kraft, *Chem. Comm.*, **17,** 2103 (1996).
6. M. Al-Talib, H. Tashtoush and N. Odeh, *Synthetic Commun.*, **20,** 1811 (1990).
7. H. Tashtoush, M. Al-Talib and N. Odeh, *Liebigs Ann. Chem.*, 291 (1992).
8. _____, *Mag. Res. Chem.*, **30,** 910 (1992).
9. H. Tashtoush, R. Subaih and M. Al-Talib, *Mag. Res. Chem.*, **35,** 549 (1997).
10. R. Huisgen, J. Sauer, H.J. Sturm and J.H. Markgraf, *Chem. Ber.*, **93,** 2107 (1960).
11. J.L. Cotter, *J. Chem. Soc.*, 5491 (1964).
12. N.A. Rodios and N.F. Alexandro, *Org. Mass. Spectrom.*, **21,** 95 (1986).
13. S.T. Abu-Orabi, M.A. Atfah, I. Jibril, A.A. Ali and F. Marii, *Gazzetta Chimica Italiana*, **122,** 29 (1992).
14. _____, *Ibid.*, **121,** 397 (1991).
15. H. Tashtoush, S. Abu Orabi, E. Taan and M. Al-Talib, *Indian J. Chem.* (in Press).
16. Khaled Abul Shendi, Master Thesis, Yarmouk University.

(*Received*: 21 September 1998; *Accepted*: 4 December 1998) AJC-1637