The Syntheses and Reactions of 3-Arylsydnone-4-carboxamide Phenylhydrazones

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Reactions of anline with 3-arylsydnone-4-carbohydroximic acid chlorides (**1**) gave the desired substitution products **5**. 3-Arylsydnone-4-carboxamide phenylhydrazones (**7**) were obtained unexpectedly by the reaction of carbohydroximic acid chlorides **1** with phenylhydrazine in suitable conditions. Compounds **7** could react with both aromatic and aliphatic aldehydes in the presence of acid catalyst to give 3-aryl-4-(1′-phenyl-5′-substituted-1′,2′,4′-triazol-3′-yl)sydnones (**11**).

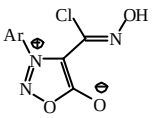
INTRODUCTION

Many sydnone compounds have been found to have pharmacological and biological activities.[1-9] In addition, 1,2,4- triazolyl derivatives are also well known to show special pharmacological activities.[10-16] It there fore should be significant to synthesize sydnone compounds having a 1,2,4-triazolyl group substituted at a suitable position. However, a sydnone ring is so sensitive to acids, alkalies and heat that reaction conditions for the preparation of sydnone derivatives seem to be limited. In our previous work, 3-arylsydnone-4-carbohydroximic acid chlorides (**1**) are stable at room temperature and are easily converted into 3-arylsydnone-4- carbonitrile oxides in polar solvent or basic aqueous solution.[17] The active nitrile oxides generated by this route could undergo cycloaddition with dipolarophiles to give 4-heterocyclic sydnones.[18-22] 3-Arylsydnone-4- carbohydroximic acid chlorides could be used to react with some nucleophiles to give the precursors which were expected to be cyclized with other reagents to give the desired 4-heterocyclic sydnones. We now report the studies on the formation of 1,2,4,-triazolyl group substituted sydnones.

RESULTS AND DISCUSSION

In previous work, 3-arylsydnone-4-carbohydroximic acid chlorides (**1**) could easily react with nucleophiles such as ammonia, hydroxylamine and hydrazinium hydroxide to give the corresponding products **2-4** which are good precusors[23-24] for the synthesis of 4-heterocyclic sydnones.

Reaction of anline with carbohydroximic acid chlorides **1** give N-phenyl-3-arylsydnone-4-carboxamide oximes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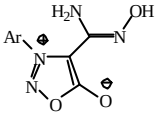
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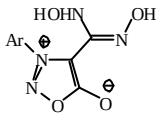
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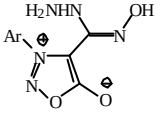
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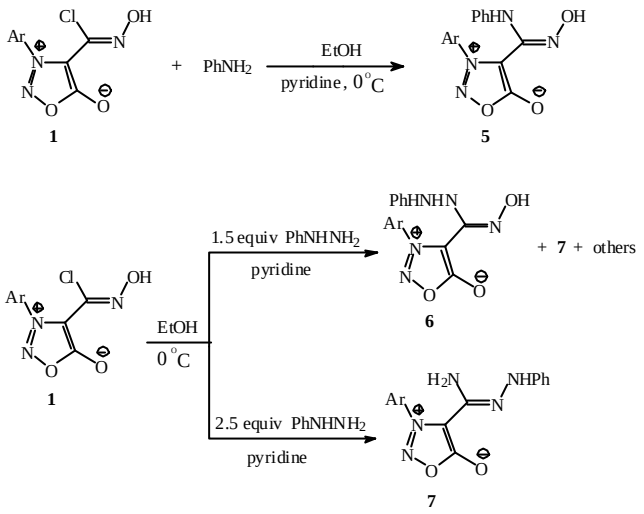
(**5**) sucessfully as common nucleophilic reagents mentioned above. In order to get the desired products, 1,2,4-triazol-3-yl- sydnones, phenylhydrazine should definitely be suggested to react with carbohydroximic acid chlorides **1** to give the desired precursors. However the substitution products were not only the expected N-phenylamino-3-arylsydnone-4- carboxamide oximes (**6**), but also 3-arylsydnone-4-carboxamide phenylhydrazones (**7**) and other compounds (Scheme I).

It is hard to explain which reaction conditions were changed including the reaction temperature, reaction time and the quantity of pyridine usage. Through several experimental tests, the problem was finally solved. If the quantity of phenylhydrazine usage was increased to 2.5 equivalent of the starting material 3-arylsydnone-4-carbohydroximic acid chloride (**1**), only product **7** would be obtained in long reaction time as shown in Scheme I.

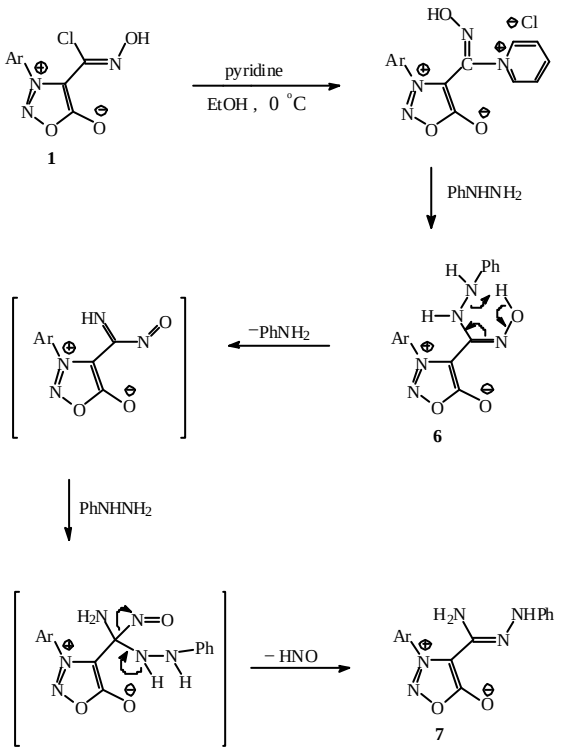
Although the reaction mechanism between compound **1** and phenylhydrazine needs further investigation, one might speculate that the mechanism would probably be similar to that of phenylosazones synthesis[25-27] and nitrosative cleavage of tertiary amine.[28-29] A molecule of anline was released and elimination of nitroxyl was proposed in the mechanism.

Scheme I

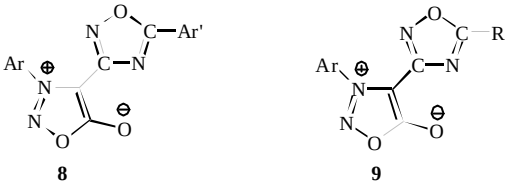


Pyridine used in the reaction was not only to remove hydrogen chloride, but also used as a nucleophilic assistant. Therefore, the mechanism might be suggested as shown in Scheme II.

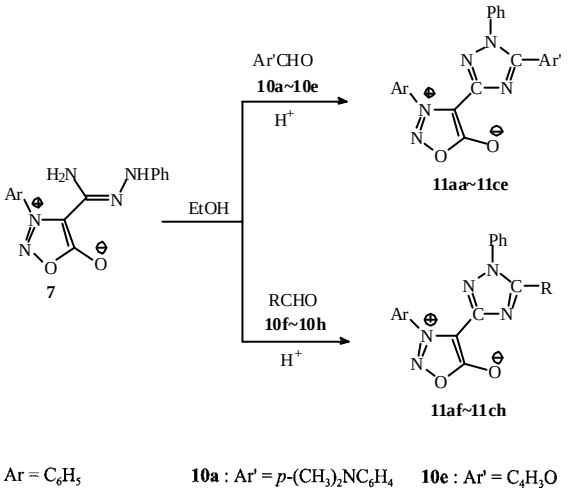
Scheme II



It is known that 3-arylsydnone-4-carboxamide oximes (**2**) and *N*-hydroxy-3-arylsydnone-4-carboxamide oximes (**3**) could cyclize with aldehydes to form 1,2,4-oxadiazol-3-ylsydnones.[23-24] The former could only react with aromatic aldehydes to give compounds **8**, but the latter reacted with both aliphatic and aromatic aldehydes sucessfully to afford compounds **8** and **9**. Therefore, 3-arylsydnone-4-carboxamide phenylhydrazones (**7**) were next suggested to react with aldehydes **10**. Indeed, the cyclizations were completely successful either with aromatic or aliphatic aldehydes under acid catalyst. The reaction products 3-aryl-4-(1′-phenyl-5′-substituted-1′,2′,4′-triazol-3′-yl)sydnones (**11**) were obtained in high yield (Scheme III). The possible cyclization mechanism is suggested in Scheme IV.



Scheme III

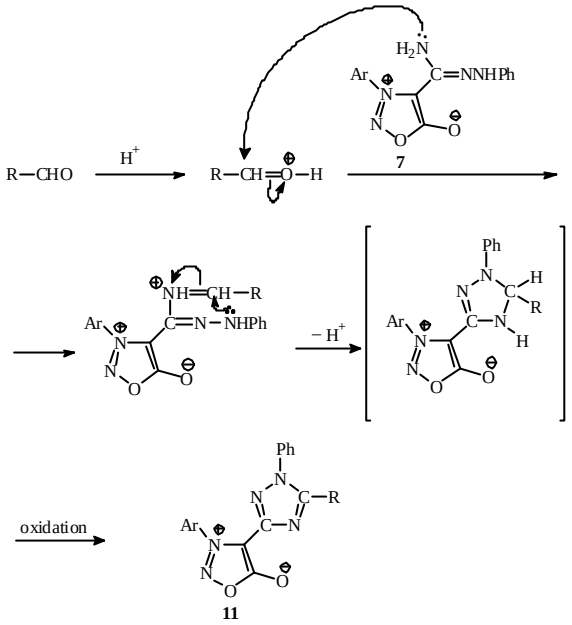


7a : Ar = C_6H_5	**10a** : Ar' = *p*-$(CH_3)_2NC_6H_4$	**10e** : Ar' = C_4H_3O
7b : Ar = *p*-$CH_3C_6H_4$	**10b** : Ar' = *m*-$CH_3OC_6H_4$	**10f** : R = CH_3CH_2
7c : Ar = *p*-$CH_3OC_6H_4$	**10c** : Ar' = C_6H_5	**10g** : R = $(CH_3)_2CH$
7d : Ar = *p*-$C_2H_5OC_6H_4$	**10d** : Ar' = *p*-$NO_2C_6H_4$	**10h** : R = $CH_3CH_2CH_2$

Among these new products, the yellow crystal **11ca** obtained was analytically pure and suitable for X-ray structure determination. The molecular structure of 3-(4′-methoxyphenyl)-4-[1′-phenyl-5′-(4″-dimethylaminophenyl)-1′,2′,4′-triazol-3′-yl] sydnone (**11ca**) is shown in Fig. 1. Details of crystal data are summarized in Tables 1-4.

In summary, the experimental results showed that 3-arylsydnone-4-carbohydroximic acid chlorides easily reacted with nucleophiles to give good precursors for syn the-

Scheme IV



siz ing 4-heterocyclic sydnones. 3-Arylsydnone-4-car boxamide phenylhydrazones (**7**) could be iso lated in a pure state through long time re ac tion of carbohydroximic acid chlo rides **1** and an ex cess amount of phenylhydrazine. The pre cur sors **7** could cyclize with both ar o matic and aliphatic al de hydes in the presence of acid cat a lyst to af ford the de sired 1,2,4-triazolyl sub sti tuted sydnones. The quan tity of sul fu ric acid us age should be con trolled ad e quately to im prove the re action rate. From sev eral ex per i men tal tests, it can be found that a small amount of sul fu ric acid cat a lyzes the re ac tion. However, a large amount of sul fu ric acid re tards the re ac tion and

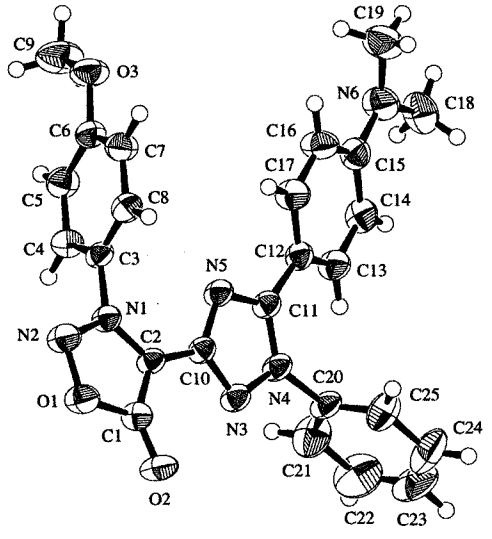


Fig. 1. Mo lec u lar struc ture of 3-(4′-methoxyphenyl)-4-[1′-phenyl-5′-(4″-dimethylaminophenyl)-1′,2′,4′-triazol-3′-yl]sydnone **(11ca)**.

Table 1. Crystal Data of Compound **11ca**

Diffractometer	Rigaku AFC7S
Formula	$C_{25}H_{22}N_6O_3$
Formula weight	454.49
Cryst system	Monoclinic
Space group	$P2_1/n$(#14)
a/Å	12.692(2)
b/Å	8.517(1)
c/Å	21.254(3)
β(deg)	96.75(1)
V/Å^3	2281.4(6)
Z	4
D_{calc}(g cm^{-3})	1.323
F_{000}	952.00
μ(Mo,K_α), cm^{-1}	0.90
Crystal size (mm)	0.40 × 0.76 × 0.96
Temperature (K)	298
Scan type	ω-2θ
$2\theta_{max}$, deg	50.0
Reflections measured	4512
Unique reflections	4307
No. observations [I>3.00σ(I)]	3087
No. variables	307
Residuals: R; Rw	0.044; 0.049
GoF	3.67

Table 2. Selected Bond Distances Å for Compound **11ca**

O(1)-N(2)	1.377(2)	O(1)-C(1)	1.419(2)
O(2)-C(1)	1.204(2)	O(3)-C(6)	1.368(2)
O(3)-C(9)	1.419(3)	N(1)-N(2)	1.313(2)
N(1)-C(2)	1.352(2)	N(1)-C(3)	1.452(2)
N(3)-N(4)	1.372(2)	N(3)-C(10)	1.328(2)
N(4)-C(11)	1.358(2)	N(4)-C(20)	1.444(2)
N(5)-C(10)	1.353(2)	N(5)-C(11)	1.333(2)
N(6)-C(15)	1.375(3)	N(6)-C(18)	1.447(3)
N(6)-C(19)	1.441(3)	C(1)-C(2)	1.414(3)
C(2)-C(10)	1.447(3)	C(3)-C(4)	1.366(3)
C(3)-C(8)	1.370(3)	C(4)-C(5)	1.385(3)
C(5)-C(6)	1.379(3)	C(6)-C(7)	1.382(3)
C(7)-C(8)	1.379(3)	C(11)-C(12)	1.459(3)
C(12)-C(13)	1.390(3)	C(12)-C(17)	1.389(3)
C(13)-C(14)	1.375(3)	C(14)-C(15)	1.400(3)
C(15)-C(16)	1.403(3)	C(16)-C(17)	1.373(3)
C(20)-C(21)	1.359(3)	C(20)-C(25)	1.364(3)
C(21)-C(22)	1.393(4)	C(22)-C(23)	1.357(5)
C(23)-C(24)	1.361(5)	C(24)-C(25)	1.390(4)

Table 3. Selected Bond Angles/deg for Compound **11ca**

N(2)-O(1)-C(1)	111.4(2)	C(6)-O(3)-C(9)	117.8(2)
N(2)-N(1)-C(2)	115.2(2)	N(2)-N(1)-C(3)	116.6(2)
C(2)-N(1)-C(3)	128.3(2)	O(1)-N(2)-N(1)	104.0(2)
N(4)-N(3)-C(10)	101.5(2)	N(3)-N(4)-C(11)	110.6(2)
N(3)-N(4)-C(20)	117.1(2)	C(11)-N(4)-C(20)	132.1(2)
C(10)-N(5)-C(11)	103.9(2)	C(15)-N(6)-C(18)	121.2(2)
C(15)-N(6)-C(19)	120.9(2)	C(18)-N(6)-C(19)	116.1(2)
O(1)-C(1)-O(2)	119.9(2)	O(1)-C(1)-C(2)	103.4(2)
O(2)-C(1)-C(2)	136.7(2)	N(1)-C(2)-C(1)	106.1(2)
N(1)-C(2)-C(10)	125.0(2)	C(1)-C(2)-C(10)	128.9(2)
N(1)-C(3)-C(4)	119.5(2)	N(1)-C(3)-C(8)	118.3(2)
C(4)-C(3)-C(8)	122.1(2)	C(3)-C(4)-C(5)	119.4(2)
C(4)-C(5)-C(6)	119.3(2)	O(3)-C(6)-C(5)	124.5(2)
O(3)-C(6)-C(7)	115.3(2)	C(5)-C(6)-C(7)	120.3(2)
C(6)-C(7)-C(8)	120.4(2)	C(3)-C(8)-C(7)	118.5(2)
N(3)-C(10)-N(5)	115.4(2)	N(3)-C(10)-C(2)	120.4(2)
N(5)-C(10)-C(2)	124.2(2)	N(4)-C(11)-N(5)	108.6(2)
N(4)-C(11)-C(12)	128.4(2)	N(5)-C(11)-C(12)	123.0(2)
C(11)-C(12)-C(13)	125.2(2)	C(11)-C(12)-C(17)	117.5(2)
C(13)-C(12)-C(17)	117.2(2)	C(12)-C(13)-C(14)	121.4(2)
C(13)-C(14)-C(15)	121.7(2)	N(6)-C(15)-C(14)	122.4(2)
N(6)-C(15)-C(16)	121.1(2)	C(14)-C(15)-C(16)	116.5(2)
C(15)-C(16)-C(17)	121.3(2)	C(12)-C(17)-C(16)	121.9(2)
N(4)-C(20)-C(21)	119.0(2)	N(4)-C(20)-C(25)	119.2(2)
C(21)-C(20)-C(25)	121.7(2)	C(20)-C(21)-C(22)	118.9(3)
C(21)-C(22)-C(23)	119.9(3)	C(22)-C(23)-C(24)	120.8(3)
C(23)-C(24)-C(25)	120.0(3)	C(20)-C(25)-C(24)	118.7(3)

reduces yields because excess acid may protonate the amino group of compound **7** and reduce its nuclophilicity.

EXPERIMENTAL SECTION

All melting points were determined on a Yanaco MP-J3 micromelting point apparatus and are uncorrected. IR spectra were recorded on a Hitachi 270-30 Infrared Spectrophotometer. Mass spectra were measured on a VG 70-250S GC/MS/MS spectrometer. ^{1}H NMR spectra were run on a Bruker AMX-200 NMR spectrometer, using TMS as internal standard. Elemental analyses were taken with a Heraeus CHN-O-Rapid Analyzer. X-ray spectra were performed on a Rigaku AFC7S diffractometer.

Starting Materials

3-Phenylsydnone-4-carbohydroximic acid chloride (**1a**), 3-(4′-methylphenyl)sydnone-4-carbohydroximic acid chloride (**1b**), 3-(4′-methoxyphenyl)sydnone-4-carbohydroximic acid chloride (**1c**) and 3-(4′-ethoxyphenyl)sydnone-4-carbohydroximic acid chloride (**1d**) were prepared according to methods in the literature.[17]

Syntheses of *N*-Phenyl-3-arylsydnone-4-carboxamide Oximes (5a-5d)

To the ice-cooled solution of aniline (121.1 mg, 1.3 mmol) in 0.5 mL 95% ethanol, pyridine (118.7 mg, 1.5 mmol) was added. The mixed base solution was stirred at 0°C. Then, the ice-cooled solution of 3-phenylsydnone-4-carbohydroximic acid chloride (239.5 mg, 1.0 mmol) in 4 mL 95% ethanol was slowly added to the above basic solution over 1 h. The reaction mixture was stirred at 0°C for 3-4 h. The yellow precipitate was collected by filtration and washed with 95% ethanol. The filtrate was poured into crushed ice, and the crude product was collected by filtration. All the solid products were combined and recrystallized from ethanol to afford 220.2 mg (74%) of *N*-phenyl-3-phenylsydnone-4-carboxamide oxime **(5a)** as yellow crystalline. The properties and analytical data of new compounds **5a-5d** are listed as follows.

N-Phenyl-3-phenylsydnone-4-carboxamide Oxime (5a)

Yellow needles; yield 74%; mp 143-144°C; IR (KBr) 3336, 3295, 1754 cm^{-1}; ^{1}H NMR (DMSO-d_6), δ10.89 (s, 1H),

Table 4. Atomic Coordinates for Compound **11ca**

Atom	X	Y	Z
O(1)	0.3941(1)	0.1327(2)	0.50327(7)
O(2)	0.4478(1)	0.2210(2)	0.60285(7)
O(3)	0.7105(1)	0.3716(2)	0.23978(8)
N(1)	0.5112(1)	0.2298(2)	0.45142(8)
N(2)	0.4253(1)	0.1428(2)	0.44348(8)
N(3)	0.6663(1)	0.3917(2)	0.59255(8)
N(4)	0.7517(1)	0.4894(2)	0.59028(8)
N(5)	0.6885(1)	0.4531(2)	0.49120(8)
N(6)	1.0735(2)	0.8857(3)	0.4225(1)
C(1)	0.4642(2)	0.2165(3)	0.5482(1)
C(2)	0.5405(2)	0.2783(3)	0.51149(9)
C(3)	0.5632(2)	0.2607(3)	0.39535(9)
C(4)	0.6451(2)	0.1669(3)	0.3822(1)
C(5)	0.6971(2)	0.2001(3)	0.3299(1)
C(6)	0.6647(2)	0.3269(3)	0.2921(1)
C(7)	0.5806(2)	0.4187(3)	0.3058(1)
C(8)	0.5295(2)	0.3865(3)	0.3581(1)
C(9)	0.7995(2)	0.2847(4)	0.2246(1)
C(10)	0.6319(2)	0.3754(3)	0.53143(9)
C(11)	0.7645(2)	0.5248(3)	0.5294(1)
C(12)	0.8460(2)	0.6223(3)	0.5059(1)
C(13)	0.9441(2)	0.6572(3)	0.5392(1)
C(14)	1.0185(2)	0.7441(3)	0.5124(1)
C(15)	0.9991(2)	0.8017(3)	0.4504(1)
C(16)	0.8999(2)	0.7660(3)	0.4172(1)
C(17)	0.8265(2)	0.6783(3)	0.4443(1)
C(18)	1.1733(2)	0.9310(3)	0.4579(1)
C(19)	1.0453(2)	0.9662(4)	0.3632(1)
C(20)	0.8051(2)	0.5441(3)	0.6500(1)
C(21)	0.8799(2)	0.4511(4)	0.6824(1)
C(22)	0.9261(3)	0.4993(5)	0.7420(2)
C(23)	0.8958(3)	0.6366(5)	0.7667(1)
C(24)	0.8218(3)	0.7298(4)	0.7336(1)
C(25)	0.7750(2)	0.6833(3)	0.6741(1)

8.52 (s, 1H), 7.69-7.31 (m, 5H), 7.19-6.90 (m, 3H), 6.57 (d, *J* = 7.3 Hz, 2H); EIMS (20 eV) *m/z* (%): 296 (M^+, 17), 266 (75), 238 (26), 220 (100), 104 (52). Anal. Calcd for $C_{15}H_{12}N_4O_3$: C, 60.81; H, 4.08; N, 18.91. Found: C, 60.80; H, 4.12; N, 18.89.

_N_-Phenyl-3-(4′-methylphenyl)sydnone-4-carboxamide Oxime (5b)

Yellow needles; yield 72%; mp 146-147 °C; IR (KBr) 3308, 3250, 1758, 1738 cm^{-1}; ^1H NMR (DMSO-d_6), δ 10.85 (s, 1H), 8.50 (s, 1H), 7.37 (d, *J* = 8.0 Hz, 2H), 7.24 (d, *J* = 8.0 Hz, 2H), 7.20-6.69 (m, 3H), 6.60 (d, *J* = 7.3 Hz, 2H), 2.39 (s, 3H); EIMS (20 eV) *m/z* (%): 310 (M^+, 6), 280 (43), 252 (20), 234 (82), 118 (100). Anal. Calcd for $C_{16}H_{14}N_4O_3$: C, 61.93; H, 4.55; N, 18.05. Found: C, 61.92; H, 4.60; N, 18.06.

_N_-Phenyl-3-(4′-methoxylphenyl)sydnone-4-carboxamide Oxime (5c)

White plates; yield 90%; mp 170-171 °C; IR (KBr) 3382, 3238, 1776 cm^{-1}; ^1H NMR (DMSO-d_6), δ 10.86 (s, 1H), 8.50 (s, 1H), 7.29 (d, *J* = 9.2 Hz, 2H), 7.13-6.90 (m, 3H), 7.07 (d, *J* = 9.2 Hz, 2H), 6.59 (d, *J* = 7.1 Hz, 2H), 3.83 (s, 3H); EIMS (20 eV) *m/z* (%): 326 (M^+, 2), 296 (14), 268 (34), 250 (64), 134 (100). Anal. Calcd for $C_{16}H_{14}N_4O_4$: C, 58.89; H, 4.32; N, 17.17. Found: C, 58.98; H, 4.33; N, 17.14.

_N_-Phenyl-3-(4′-ethoxyphenyl)sydnone-4-carboxamide Oxime (5d)

White plates; yield 92%; mp 155-156 °C; IR (KBr) 3384, 3208, 1760 cm^{-1}; ^1H NMR (DMSO-d_6), δ 10.86 (s, 1H), 8.50 (s, 1H), 7.26 (d, *J* = 9.2 Hz, 2H), 7.13-6.83 (m, 3H), 7.04 (d, *J* = 9.2 Hz, 2H), 6.58 (d, *J* = 7.3 Hz, 2H), 4.11 (q, *J* = 6.9 Hz, 2H), 1.35 (t, *J* = 6.9 Hz, 3H); EIMS (20 eV) *m/z* (%): 340 (M^+, 10), 310 (11), 282 (23), 264 (45), 148 (100). Anal. Calcd for $C_{17}H_{16}N_4O_4$: C, 60.00; H, 4.74; N, 16.46. Found: C, 60.00; H, 4.75; N, 16.52.

Syntheses of 3-Arylsydnone-4-carboxamide Phenylhydrazones (7a-7d)

To an ice cooled solution of phenylhydrazine (248.7 mg, 2.3 mmol) in 95% ethanol (0.5 mL), pyridine (197.8 mg, 2.5 mmol) was added. The mixed basic solution was stirred at 0 °C. An ice-cooled solution of 3-phenylsydnone-4-carbohydroximic acid chloride (239.5 mg, 1.0 mmol) in 95% ethanol (4 mL) was slowly added to the above basic solution over 1 h. The reaction mixture was stirred for further 2-3 h at 0°C, then kept stirring at room temperature until the reaction was completed. The resulting dark red solution was concentrated and filtered. The obtained red solid was recrystallized from 95% ethanol to afford 3-phenylsydnone-4-carboxamide phenylhydrazone (**7a**, 104.1 mg, 35%).

3-Phenylsydnone-4-carboxamide Phenylhydrazone (7a)

Red needles; yield 35%; mp 171-172 °C; IR (KBr) 3460, 3364, 3320, 1722 cm^{-1}; ^1H NMR (DMSO-d_6), δ 8.38 (s, 1H), 7.73-7.67 (m, 5H), 6.90 (t, *J* = 8.1 Hz, 2H), 6.53 (tt, *J* = 8.1, 1.4 Hz, 1H), 6.21 (dd, *J* = 8.1, 1.4 Hz, 2H), 5.95 (s, 2H); EIMS (70 eV) *m/z* (%): 295 (M^+, 25), 265 (33), 237 (48), 104 (47), 92(100). Anal. Calcd for $C_{15}H_{13}N_5O_2$: C, 61.01; H, 4.44; N, 23.72. Found: C, 60.99; H, 4.48; N, 23.72.

3-(4′-Methylphenyl)sydnone-4-carboxamide Phenylhydrazone (7b)

Red needles; yield 40%; mp 167-168 °C; IR (KBr) 3432, 3332, 3304, 1724 cm^{-1}; ^1H NMR (DMSO-d_6), δ 8.36 (s,

1H), 7.60 (d, *J* = 8.6 Hz, 2H), 7.46 (d, *J* = 8.6 Hz, 2H), 6.95 (t, *J* = 7.3 Hz, 2H), 6.58 (t, *J* = 7.3 Hz, 1H), 6.24 (d, *J* = 7.3 Hz, 2H), 5.93 (s, 2H), 2.47 (s, 3H); EIMS (20 eV) *m/z* (%): 309 (M^+, 100), 279 (92), 251 (79), 118 (47), 92 (51). Anal. Calcd for $C_{16}H_{15}N_5O_2$: C, 62.13; H, 4.89; N, 22.64. Found: C, 62.04; H, 4.93; N, 22.60.

3-(4′-Methoxyphenyl)sydnone-4-carboxamide Phenylhydrazone (7c)

Red needles; yield 50%; mp 189-190 °C; IR (KBr) 3436, 3332, 3308, 1726, 1716 cm^{-1}; ^1H NMR (DMSO-d_6), δ 8.37 (s, 1H), 7.65 (d, *J* = 9.1 Hz, 2H), 7.18 (d, *J* = 9.1 Hz, 2H), 6.95 (t, *J* = 7.2 Hz, 2H), 6.55 (t, *J* = 7.2 Hz, 1H), 6.31 (d, *J* = 7.2 Hz, 2H), 5.94 (s, 2H), 3.87 (s, 3H); EIMS (70 eV) *m/z* (%): 325 (M^+, 59), 295 (43), 267 (45), 134 (100), 92 (94). Anal. Calcd for $C_{16}H_{15}N_5O_3$: C, 59.07; H, 4.65; N, 21.53. Found: C, 58.93; H, 4.66; N, 21.48.

3-(4′-Ethoxyphenyl)sydnone-4-carboxamide Phenylhydrazone (7d)

Red needles; yield 52%; mp 180-182 °C; IR (KBr) 3448, 3348, 3324, 1722 cm^{-1}; ^1H NMR (DMSO-d_6), δ 8.37 (s, 1H), 7.63 (d, *J* = 9.0 Hz, 2H), 7.16 (d, *J* = 9.0 Hz, 2H), 6.95 (t, *J* = 7.2 Hz, 2H), 6.55 (t, *J* = 7.2 Hz, 1H), 6.33 (d, *J* = 7.2 Hz, 2H), 5.95 (s, 2H), 4.14 (q, *J* = 6.9 Hz, 2H), 1.38 (t, *J* = 6.9 Hz, 3H); EIMS (20 eV) *m/z* (%): 339 (M^+, 53), 309 (34), 281 (31), 148 (82), 92 (100). Anal. Calcd for $C_{17}H_{17}N_5O_3$: C, 60.17; H, 5.05; N, 20.64. Found: C, 59.94; H, 5.05; N, 20.43.

Syntheses of 3-Aryl-4-(1′-phenyl -5′-aryl-1′,2′,4′-triazol-3′-yl)sydnones (11aa-11ce)

Typical procedures for the syntheses of sydnone derivatives **11aa-11ce** were as follows:

To a solution of 3-phenylsydnone-4-carboxamide phenylhydrazone (**7a**, 118.1 mg, 0.4 mmol) and 4-(dimethylamino)benzaldehyde (**10a**, 71.6 mg, 0.48 mmol) in 95% ethanol (3 mL), a drop of sulfuric acid (98%) was added as catalytic agent. The reaction mixture was stirred at room temperature for 4 days. The precipitation solid was collected by filtration and then recrystallized from dichloromethane-ethanol to afford 3-phenyl-4-[1′-phenyl-5′-(4″-dimethylaminophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (**11aa**, 128.0 mg, 75%).

3-Phenyl-4-[1′-phenyl-5′-(4″-dimethylaminophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11aa)

Yellow crystals from CH_2Cl_2/EtOH; yield 75%; mp 212-213 °C; IR (KBr) 1766 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.91-7.64 (m, 5H), 7.56-7.28 (m, 5H), 7.04 (d, *J* = 9.0 Hz, 2H), 6.58 (d, *J* = 9.0 Hz, 2H), 2.89 (s, 6H); EIMS (70 eV) *m/z* (%): 424 (M^+, 23), 366 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 19). Anal. Calcd for $C_{24}H_{20}N_6O_2$: C, 67.91; H, 4.75; N, 19.80. Found: C, 67.77; H, 4.80; N, 19.64.

3-(4′-Methylphenyl)-4-[1′-phenyl-5′-(4″-dimethylaminophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11ba)

Yellow crystals from CH_2Cl_2/EtOH; yield 80%; mp 191-192 °C; IR (KBr) 1768 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.71 (d, *J* = 8.4 Hz, 2H), 7.56-7.30 (m, 7H), 7.07 (d, *J* = 9.0 Hz, 2H), 6.59 (d, *J* = 9.0 Hz, 2H), 2.90 (s, 6H), 2.44 (s, 3H); EIMS (30 eV) *m/z* (%): 438 (M^+, 13), 380 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 7). Anal. Calcd for $C_{25}H_{22}N_6O_2$: C, 68.48; H, 5.06; N, 19.17. Found: C, 68.21; H, 5.16; N, 19.08.

3-(4′-Methoxyphenyl)-4-[1′-phenyl-5′-(4″-dimethylaminophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11ca)

Yellow crystals from CH_2Cl_2/EtOH; yield 87%; mp 235-236 °C; IR (KBr) 1770 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.77 (d, *J* = 9.0 Hz, 2H), 7.56-7.35 (m, 5H), 7.18 (d, *J* = 9.0 Hz, 2H), 7.10 (d, *J* = 8.9 Hz, 2H), 6.60 (d, *J* = 8.9 Hz, 2H), 3.86 (s, 3H), 2.90 (s, 6H); EIMS (30 eV) *m/z* (%): 454 (M^+, 12), 396 (M^+-NO-CO, 100). Anal. Calcd for $C_{25}H_{22}N_6O_3$: C, 66.07; H, 4.88; N, 18.49. Found: C, 65.98; H, 4.93; N, 18.43.

3-(4′-Ethoxyphenyl)-4-[1′-phenyl-5′-(4″-dimethylaminophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11da)

Yellow crystals from CH_2Cl_2/EtOH; yield 85%; mp 195-196 °C; IR (KBr) 1770 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.75 (d, *J* = 8.9 Hz, 2H), 7.56-7.32 (m, 5H), 7.15 (d, *J* = 8.9 Hz, 2H), 7.09 (d, *J* = 9.0 Hz, 2H), 6.60 (d, *J* = 9.0 Hz, 2H), 4.14 (q, *J* = 6.9 Hz, 2H), 2.91 (s, 6H), 1.36 (t, *J* = 6.9 Hz, 3H); EIMS (70 eV) *m/z* (%): 468 (M^+, 17), 410 (M^+-NO-CO, 100). Anal. Calcd for $C_{26}H_{24}N_6O_3$: C, 66.65; H, 5.16; N, 17.94. Found: C, 66.48; H, 5.24; N, 17.86.

3-Phenyl-4-[1′-phenyl-5′-(3″-methoxyphenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11ab)

White crystals from CH_2Cl_2/EtOH; yield 75%; mp 199-200 °C; IR (KBr) 1770 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.92-7.65 (m, 5H), 7.56-7.18 (m, 6H), 7.05-6.74 (m, 3H), 3.61 (s, 3H); EIMS (30 eV) *m/z* (%): 411 (M^+, 15), 353 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 18). Anal. Calcd for $C_{23}H_{17}N_5O_3$: C, 67.15; H, 4.16; N, 17.02. Found: C, 67.12; H, 4.22; N, 17.02.

3-(4′-Methylphenyl)-4-[1′-phenyl-5′-(3″-methoxyphenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11bb)

Yellow crystals from CH_2Cl_2/EtOH; yield 78%; mp 194-195 °C; IR (KBr) 1768 (ν C=O) cm^{-1}; ^1H NMR (DMSO-

d$_6$), δ 7.73 (d, *J* = 8.4 Hz, 2H), 7.57-7.18 (m, 8H), 6.97-6.79 (m, 3H), 3.62 (s, 3H), 2.43 (s, 3H); EIMS (30 eV) *m/z* (%): 425 (M^+, 14), 367 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 20). Anal. Calcd for $C_{24}H_{19}N_5O_3$: C, 67.76; H, 4.50; N, 16.46. Found: C, 67.53; H, 4.55; N, 16.35.

3-(4′-Methoxyphenyl)-4-[1′-phenyl-5′-(3″-methoxyphenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11cb)

Yellow crystals from CH_2Cl_2/EtOH; yield 81%; mp 208-209 °C; IR (KBr) 1766 (ν C=O) cm^{-1}; ^{1}H NMR (DMSO-*d*$_6$), δ 7.79 (d, *J* = 9.0 Hz, 2H), 7.57-7.35 (m, 5H), 7.27-7.14 (m, 3H), 7.03-6.76 (m, 3H), 3.86 (s, 3H), 3.62 (s, 3H); EIMS (30 eV) *m/z* (%): 441 (M^+, 6), 383 (M^+-NO-CO, 100), 107 ($CH_3OC_6H_4^+$, 4). Anal. Calcd for $C_{24}H_{19}N_5O_4$: C, 65.30; H, 4.34; N, 15.87. Found: C, 65.11; H, 4.40; N, 15.89.

3-(4′-Ethoxyphenyl)-4-[1′-phenyl-5′-(3″-methoxyphenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11db)

Yellow crystals from CH_2Cl_2/EtOH; yield 80%; mp 214-215 °C; IR (KBr) 1766 (ν C=O) cm^{-1}; ^{1}H NMR (Acetone-*d*$_6$ + DMSO-*d*$_6$), δ 7.78 (d, *J* = 9.0 Hz, 2H), 7.58-7.42 (m, 5H), 7.27-7.14 (m, 3H), 7.04-6.93 (m, 3H), 4.19 (q, *J* = 6.9 Hz, 2H), 3.68 (s, 3H), 1.41 (t, *J* = 6.9 Hz, 3H); EIMS (30 eV) *m/z* (%): 455 (M^+, 5), 397 (M^+-NO-CO, 100), 121 ($C_2H_5OC_6H_4^+$, 5). Anal. Calcd for $C_{25}H_{21}N_5O_4$: C, 65.93; H, 4.65; N, 15.38. Found: C, 65.85; H, 4.74; N, 15.17.

3-Phenyl-4-(1′,5′-diphenyl-1′,2′,4′-triazol-3′-yl)sydnone (11ac)

White needles from CH_3COCH_3/EtOH; yield 74%; mp 209-210 °C; IR (KBr) 1761 (ν C=O) cm^{-1}; ^{1}H NMR (Acetone-*d*$_6$), δ 7.92-7.63 (m, 5H), 7.58-7.38 (m, 5H), 7.37-7.31 (m, 5H); EIMS (30 eV) *m/z* (%): 381(M^+, 9), 323 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 4). Anal. Calcd for $C_{22}H_{15}N_5O_2$: C, 69.28; H, 3.96; N, 18.36. Found: C, 69.21; H, 4.14; N, 18.18.

3-(4′-Methylphenyl)-4-(1′,5′-diphenyl-1′,2′,4′-triazol-3′-yl)-sydnone (11bc)

White needles from CH_3COCH_3/EtOH; yield 70%; mp 210-210.5 °C; IR (KBr) 1770 (ν C=O) cm^{-1}; ^{1}H NMR (DMSO-*d*$_6$), δ 7.73 (d, *J* = 8.5 Hz, 2H), 7.54-7.44 (m, 5H), 7.41-7.24 (m, 7H), 2.43 (s, 3H); EIMS (30 eV) *m/z* (%): 395 (M^+, 11), 337 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 19). Anal. Calcd for $C_{23}H_{17}N_5O_2$: C, 69.86; H, 4.33; N, 17.71. Found: C, 69.84; H, 4.36; N, 17.66.

3-(4′-Methoxyphenyl)-4-(1′,5′-diphenyl-1′,2′,4′-triazol-3′-yl)sydnone (11cc)

White needles from CH_3COCH_3/EtOH; yield 85%; mp 213-214 °C; IR (KBr) 1774 (ν C=O) cm^{-1}; ^{1}H NMR (DMSO-*d*$_6$), δ 7.78 (d, *J* = 9.0 Hz, 2H), 7.55-7.42 (m, 5H), 7.39-7.26 (m, 5H), 7.18 (d, *J* = 9.0 Hz, 2H), 3.86 (s, 3H); EIMS (70 eV) *m/z* (%): 411 (M^+, 4), 353 (M^+-NO-CO, 100), 107 ($CH_3OC_6H_4^+$, 6). Anal. Calcd for $C_{23}H_{17}N_5O_3$: C, 67.15; H, 4.16; N, 17.02. Found: C, 66.91; H, 4.25; N, 16.95.

3-(4′-Ethoxyphenyl)-4-(1′,5′-diphenyl-1′,2′,4′-triazol-3′-yl)-sydnone (11dc)

White needles from CH_3COCH_3/EtOH; yield 88%; mp 197-198 °C; IR (KBr) 1766 (ν C=O) cm^{-1}; ^{1}H NMR (DMSO-*d*$_6$), δ 7.76 (d, *J* = 9.0 Hz, 2H), 7.55-7.42 (m, 5H), 7.39-7.23 (m, 5H), 7.15 (d, *J* = 9.0 Hz, 2H), 4.14 (q, *J* = 6.9 Hz, 2H), 1.36 (t, *J* = 6.9 Hz, 3H); EIMS (70 eV) *m/z* (%): 425 (M^+, 3), 367 (M^+-NO-CO, 100), 121 ($C_2H_5OC_6H_4^+$, 3). Anal. Calcd for $C_{24}H_{19}N_5O_3$: C, 67.76; H, 4.50; N, 16.46. Found: C, 67.71; H, 4.53; N, 16.41.

3-Phenyl-4-[1′-phenyl-5′-(4″-nitrophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11ad)

Yellow needles from CH_3COCH_3/EtOH; yield 72%; mp 195-196 °C; IR (KBr) 1770 (ν C=O), 1524, 1340 (ν NO_2) cm^{-1}; ^{1}H NMR (Acetone-*d*$_6$), δ 8.23 (d, *J* = 9.1 Hz, 2H), 7.93-7.67 (m, 7H), 7.62-7.35 (m, 5H); EIMS (30 eV) *m/z* (%): 426 (M^+, 8), 368 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 3). Anal. Calcd for $C_{22}H_{14}N_6O_4$: C, 61.97; H, 3.31; N, 19.71. Found: C, 61.76; H, 3.47; N, 19.58.

3-(4′-Methylphenyl)-4-[1′-phenyl-5′-(4″-nitrophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11bd)

Yellow needles from CH_3COCH_3/EtOH; yield 76%; mp 163-164 °C; IR (KBr) 1770 (ν C=O), 1524, 1344 (ν NO_2) cm^{-1}; ^{1}H NMR (Acetone-*d*$_6$), δ 8.23 (d, *J* = 8.8 Hz, 2H), 7.75 (d, *J* = 8.8 Hz, 2H), 7.68-7.49 (m, 7H), 7.42 (d, *J* = 8.8 Hz, 2H), 2.49 (s, 3H); EIMS (30 eV) *m/z* (%): 440 (M^+, 5), 382 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 6). Anal. Calcd for $C_{23}H_{16}N_6O_4$: C, 62.73; H, 3.66; N, 19.08. Found: C, 62.54; H, 3.75; N, 19.04.

3-(4′-Methoxyphenyl)-4-[1′-phenyl-5′-(4″-nitrophenyl)-1′,2′,4′-triazol-3′-yl]sydnone (11cd)

Yellow needles from CH_3COCH_3/EtOH; yield 83%; mp 239-240 °C; IR (KBr) 1772 (ν C=O), 1528, 1346 (ν NO_2) cm^{-1}; ^{1}H NMR (DMSO-*d*$_6$), δ 8.23 (d, *J* = 9.0 Hz, 2H), 7.79 (d, *J* = 9.0 Hz, 2H), 7.61-7.48 (m, 5H), 7.47 (d, *J* = 9.0 Hz, 2H), 7.17 (d, *J* = 9.0 Hz, 2H), 3.86 (s, 3H); EIMS (70 eV) *m/z* (%): 456 (M^+, 4), 398 (M^+-NO-CO, 100), 107 ($CH_3OC_6H_4^+$, 9). Anal. Calcd for $C_{23}H_{16}N_6O_5$: C, 60.53; H, 3.53; N, 18.41. Found: C, 60.37; H, 3.62; N, 18.39.

3-Phenyl-4-[1′-phenyl-5′-(furan-2″-yl)-1′,2′,4′-triazol-3′-yl] sydnone (11ae)

Yellow needles from CH_3COCH_3/EtOH; yield 90%; mp 257-258 °C; IR (KBr) 1786, 1768 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.86-7.64 (m, 5H), 7.60-7.42 (m, 6H), 6.58-6.49 (m, 2H); EIMS (30 eV) *m/z* (%): 371 (M^+, 15), 313 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 35). Anal. Calcd for $C_{20}H_{13}N_5O_3$: C, 64.68; H, 3.53; N, 18.86. Found: C, 64.49; H, 3.59; N, 18.79.

3-(4′-Methylphenyl)-4-[1′-phenyl-5′-(furan-2″-yl)-1′,2′,4′-triazol-3′-yl]sydnone (11be)

White needles from CH_3COCH_3/EtOH; yield 91%; mp 208-209 °C; IR (KBr) 1768, 1748 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.71 (d, *J* = 8.8 Hz, 2H), 7.61-7.42 (m, 8H), 6.58-6.48 (m, 2H), 2.47 (s, 3H); EIMS (30 eV) *m/z* (%): 385 (M^+, 15), 327 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 31). Anal. Calcd for $C_{21}H_{15}N_5O_3$: C, 65.45; H, 3.92; N, 18.17. Found: C, 65.39; H, 4.05; N, 17.98.

3-(4′-Methoxyphenyl)-4-[1′-phenyl-5′-(furan-2″-yl)-1′,2′,4′-triazol-3′-yl]sydnone (11ce)

White needles from CH_3COCH_3/EtOH; yield 85%; mp 212-213 °C; IR (KBr) 1790, 1774 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.74 (d, *J* = 9.1 Hz, 2H), 7.60-7.34 (m, 6H), 7.16 (d, *J* = 9.1 Hz, 2H), 6.59-6.49 (m, 2H), 3.90 (s, 3H); EIMS (70 eV) *m/z* (%): 401 (M^+, 6), 343 (M^+-NO-CO, 100), 107 ($CH_3OC_6H_4^+$, 3). Anal. Calcd for $C_{21}H_{15}N_5O_4$: C, 62.84; H, 3.77; N, 17.45. Found: C, 62.73; H, 3.82; N, 17.37.

Syntheses of 3-Aryl-4-(1′-phenyl-5′-alkyl-1′,2′,4′-triazol-3′-yl)sydnones (11af-11ch)

Typical procedures for the syntheses of sydnone derivatives **11af-11ch** were as follows:

3-Phenyl-4-(1′-phenyl-5′-ethyl-1′,2′,4′-triazol-3′-yl)-sydnone (**11af**) was prepared in 73% yield (97.2 mg, 2.9 mmol) from a ethanol solution of 3-phenylsydnone-4-carboxamide phenylhydrazone (**7a**, 118.1 mg, 0.4 mmol) and propionaldehyde (**10f**, 185.9 mg, 3.2 mmol) in the presence of sulfuric acid catalyst by a procedure similar to that for compound **11aa**.

3-Phenyl-4-(1′-phenyl-5′-ethyl-1′,2′,4′-triazol-3′-yl)sydnone (11af)

White needles from EtOH; yield 73%; mp 212-213 °C; IR (KBr) 1766 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.85-7.68 (m, 5H), 7.63-7.39 (m, 5H), 2.70 (q, *J* = 7.5 Hz, 2H), 1.04 (t, *J* = 7.5 Hz, 3H); EIMS (30 eV) *m/z* (%): 333 (M^+, 13), 275 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 25). Anal. Calcd for $C_{18}H_{15}N_5O_2$: C, 64.86; H, 4.54; N, 21.01. Found: C, 64.73; H, 4.57; N, 20.99.

3-(4′-Methylphenyl)-4-(1′-phenyl-5′-ethyl-1′,2′,4′-triazol-3′-yl)sydnone (11bf)

White needles from EtOH; yield 78%; mp 202-203 °C; IR (KBr) 1762 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.66 (d, *J* = 8.3 Hz, 2H), 7.52-7.47 (m, 5H), 7.43 (d, *J* = 8.3 Hz, 2H), 2.71 (q, *J* = 7.4 Hz, 2H), 2.42 (s, 3H), 1.05 (t, *J* = 7.4 Hz, 3H); EIMS (30 eV) *m/z* (%): 347 (M^+, 10), 289 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 23). Anal. Calcd for $C_{19}H_{17}N_5O_2$: C, 65.70; H, 4.93; N, 20.16. Found: C, 65.42; H, 4.97; N, 20.09.

3-(4′-Methoxyphenyl)-4-(1′-phenyl-5′-ethyl-1′,2′,4′-triazol-3′-yl)sydnone (11cf)

White needles from EtOH; yield 72%; mp 193-194 °C; IR (KBr) 1776, 1766 (ν C=O) cm^{-1}; ^1H NMR (DMSO-d_6), δ 7.71 (d, *J* = 9.0 Hz, 2H), 7.61-7.42 (m, 5H), 7.15 (d, *J* = 9.0 Hz, 2H), 3.85 (s, 3H), 2.72 (q, *J* = 7.5 Hz, 2H), 1.07 (t, *J* = 7.5 Hz, 3H); EIMS (30 eV) *m/z* (%): 363 (M^+, 5), 305 (M^+-NO-CO, 100), 107 ($CH_3OC_6H_4^+$, 2). Anal. Calcd for $C_{19}H_{17}N_5O_3$: C, 62.80; H, 4.72; N, 19.27. Found: C, 62.64; H, 4.75; N, 19.19.

3-Phenyl-4-(1′-phenyl-5′-isopropyl-1′,2′,4′-triazol-3′-yl)-sydnone (11ag)

White needles from CH_3COCH_3/EtOH; yield 90%; mp 158-159 °C; IR (KBr) 1794, 1774 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.82-7.64 (m, 5H), 7.61-7.39 (m, 5H), 3.25-2.96 (m, 1H), 1.10 (d, *J* = 6.8 Hz, 6H); EIMS (30 eV) *m/z* (%): 347 (M^+, 15), 289 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 4). Anal. Calcd for $C_{19}H_{17}N_5O_2$: C, 65.70; H, 4.93; N, 20.16. Found: C, 65.64; H, 4.96; N, 20.14.

3-(4′-Methylphenyl)-4-(1′-phenyl-5′-isopropyl-1′,2′,4′-triazol-3′-yl)sydnone (11bg)

White plates from CH_3COCH_3/EtOH; yield 94%; mp 147-148 °C; IR (KBr) 1778 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.65 (d, *J* = 8.5 Hz, 2H), 7.56-7.40 (m, 7H), 3.26-2.97 (m, 1H), 2.46 (s, 3H), 1.12 (d, *J* = 6.8 Hz, 6H); EIMS (30 eV) *m/z* (%): 361 (M^+, 12), 303 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 12). Anal. Calcd for $C_{20}H_{19}N_5O_2$: C, 66.47; H, 5.30; N, 19.38. Found: C, 66.42; H, 5.34; N, 19.36.

3-(4′-Methoxyphenyl)-4-(1′-phenyl-5′-isopropyl-1′,2′,4′-triazol-3′-yl)sydnone (11cg)

White needles from CH_3COCH_3/EtOH; yield 92%; mp 149-150 °C; IR (KBr) 1792, 1776 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.70 (d, *J* = 9.1 Hz, 2H), 7.62-7.43 (m, 5H),

7.15 (d, J = 9.1 Hz, 2H), 3.91 (s, 3H), 3.27-2.98 (m, 1H), 1.14 (d, J = 6.8 Hz, 6H); EIMS (30 eV) *m/z* (%): 377 (M^+, 7), 319 (M^+-NO-CO, 100), 107 ($CH_3OC_6H_4^+$, 3). Anal. Calcd for $C_{20}H_{19}N_5O_3$: C, 63.65; H, 5.07; N, 18.56. Found: C, 63.44; H, 5.06; N, 18.49.

3-Phenyl-4-(1′-phenyl-5′-propyl-1′,2′,4′-triazol-3′-yl)-sydnone (11ah)

Yellow needles from CH_3COCH_3/EtOH; yield 74%; mp 167-168 °C; IR (KBr) 1788, 1776 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.86-7.63 (m, 5H), 7.62-7.42 (m, 5H), 2.71 (t, J = 7.4 Hz, 2H), 1.78-1.41 (m, 2H), 0.80 (t, J = 7.2 Hz, 3H); EIMS (30 eV) *m/z* (%): 347 (M^+, 10), 289 (M^+-NO-CO, 100), 77 ($C_6H_5^+$, 2). Anal. Calcd for $C_{19}H_{17}N_5O_2$: C, 65.70; H, 4.93; N, 20.16. Found: C, 65.63; H, 4.96; N, 20.16.

3-(4′-Methylphenyl)-4-(1′-phenyl-5′-propyl-1′,2′,4′-triazol-3′-yl)sydnone (11bh)

White plates from CH_3COCH_3/EtOH; yield 70%; mp 145-146 °C; IR (KBr) 1762 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.66 (d, J = 8.5 Hz, 2H), 7.56-7.41 (m, 7H), 2.72 (t, J = 7.4 Hz, 2H), 2.46 (s, 3H), 1.78-1.44 (m, 2H), 0.81 (t, J = 7.0 Hz, 3H); EIMS (30 eV) *m/z* (%): 361 (M^+, 7), 303 (M^+-NO-CO, 100), 91 ($CH_3C_6H_4^+$, 11). Anal. Calcd for $C_{20}H_{19}N_5O_2$: C, 66.47; H, 5.30; N, 19.38. Found: C, 66.46; H, 5.27; N, 19.39.

3-(4′-Methoxyphenyl)-4-(1′-phenyl-5′-propyl-1′,2′,4′-triazol-3′-yl)sydnone (11ch)

White plates from CH_3COCH_3/EtOH; yield 86%; mp 169-170 °C; IR (KBr) 1790, 1780 (ν C=O) cm^{-1}; ^1H NMR (Acetone-d_6), δ 7.71 (d, J = 9.1 Hz, 2H), 7.62-7.45 (m, 5H), 7.16 (d, J = 9.1 Hz, 2H), 3.92 (s, 3H), 2.73 (t, J = 7.4 Hz, 2H), 1.76-1.39 (m, 2H), 0.82 (t, J = 7.2 Hz, 3H); EIMS (30 eV) *m/z* (%): 377 (M^+, 5), 319 (M^+-NO-CO, 100), 107 ($CH_3OC_6H_4^+$, 1). Anal. Calcd for $C_{20}H_{19}N_5O_3$: C, 63.65; H, 5.07; N, 18.56. Found: C, 63.44; H, 5.06; N, 18.49.

Crystallography

Crystal data for compound **11ca** are summarized in Table 1. Selected bond distances and bond angles are listed in Table 2 and Table 3, respectively. Atomic coordinates are given in Table 4.

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Key Words

Sydnones; Nitrile oxides; Carbohydroximic acid chlorides; 1,2,4-Triazoles; Carboxamide oximes; 3-Arylsydnone-4-carboxamide phenylhydrazones; 3-Aryl-4-(1-phenyl-5-substituted-1,2,4-triazol-3-yl)-sydnones.

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