Fatty acid hydrazides in Organic Synthesis: Novel Synthesis of 6-alkyl-3-aryl-5-imino-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile and 6-alkyl -3-aryl-5,7-dioxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile

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Abstract

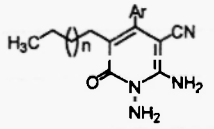
1,2- Diazepinone derivatives **5a-o** and **7a-c** were synthesized from the reaction of α,β-unsaturated nitriles **2a-d** and **6a-c** with caproic, caprylic, capric and lauric acid hydrazides respectively.

Introduction

During the last century, the production and utilization of oils, fats and their derivatives grow both in size and diversity in the industrial field[1,2] . There has been a competition between oleochemicals and petrochemicals . More recently, some fatty acid derivatives have shown insecticidal and antimicrobial properties. Moreover, α,β-unsaturated nitriles are versatile reagents which have been extensively utilized in heterocycles synthesis[3-5] . The reactivity of α,β- unsaturated nitriles towards fatty acid hydrazides have never been reported before .

In connection with the ongoing work aimed to the synthesis of fused heterocycles and study the reactivity of fatty acid hydrrazide towards carbon carbon double bond derivatives as electron-defficient alkenes.

Trials to prepare of N-amino-2-pyridones **I** by using fatty acid hydrazides was failed 1,2-diazipinone derivatives **5,7** were isolated instead .



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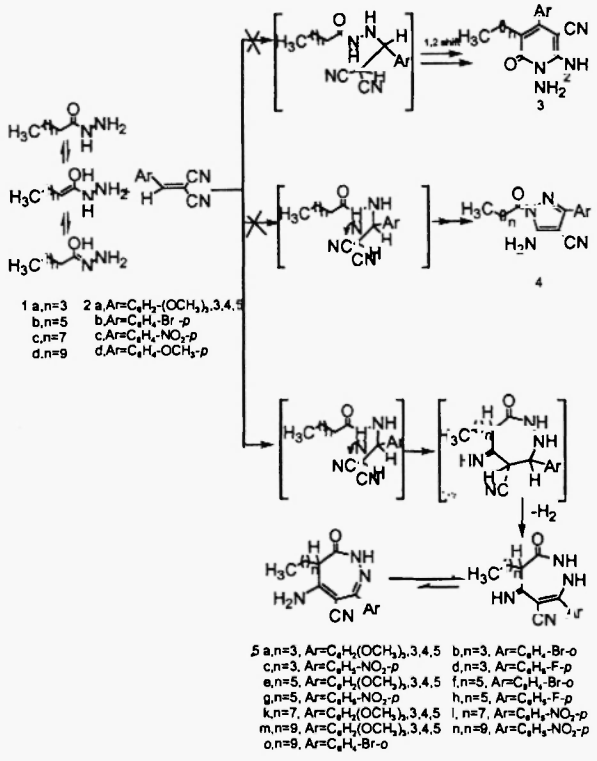
RESULTS AND DISCUSIONS

Compounds **1 a-d** namely caproic, caprylic, capric and lauric acid hydrazides reacted with benzylidene malononitriles derivatives **2a-d** which are easily prepared according to a knoevengel condensation[6-9]. The reaction is easily performed in ethanol . The nature of the substituent present in the benzene ring of benzylidene malononitrile has a little effect on the reaction. The reaction may be assumed to proceed as shown in Scheme 1, whish is assumed to involve the Michael addition of 1 to 2. The resulting adduct undergoes cyclization *in situ* by nucleophilic attack ,of CH_2CO that act as carbon acid[10], at the cyano group to give the six membered ring which on aromatization gives the N-

amino-2-pyridone **3** .However, as was previously reported[13], the characterization of the isolated product disagreed with the characterization of N-amino-2 aminopyridone **3** . The IR spectrum of the isolated product displayed characteristic absorption band at about 1650-1670 cm^{-1} which can be assigned to the carbonyl group, and absence of amino group signal in the ^{1}nmr ($CDCl_3$) (in the region δ5-6 ppm which is normally expected) with appearing of one singlet at region δ9-10ppm corresponding to NH acidic (D_2O exchangeable). Moreover, the ^{13}C nmr ($CDCl_3$) spectrum of the isolated product showed signal assigned to carbon atom of the carbonyl group that resonated at the region δ175-180ppm region corresponding to the isolated products, and does not belong the carbonyl carbon of N-amino 2-pyridone, normally seen in δ155-160 ppm[11]
region.

Also, the postulation of formation of N-pyrazolyl derivatives[12] can be eliminated since the ^{13}C nmr spectrum that showed resonance at δ140-150 ppm corresponding to the N C Ogroup is absent in the afforded product .

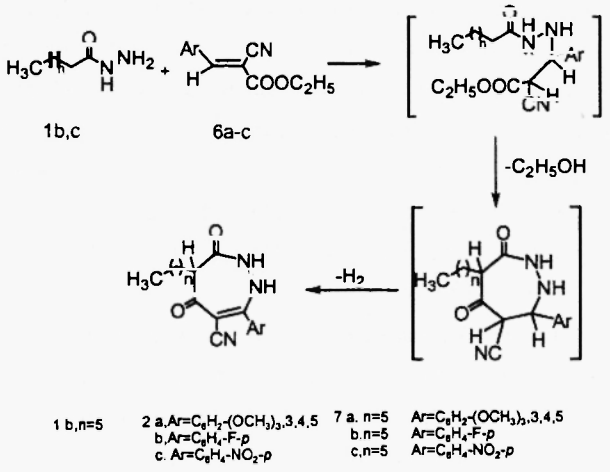


Scheme 1

Cyclization of Michael adduct to a seven membered diazapine ring is possible and must be favored by the high nucleophilic character of CH_2CO due to the presence of base , that act as nucleophile, with respect to the CO-NH group . We suggest that the isolated product is 1,2- diazapinone, and this was supported by analytical and spectral data . The 1Hnmr spectrum of 6-decyl-5-imino-7-oxo-3-(4-nitrophenyl)-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile **5n** as an example showed triplet at δ2.6 corresponding to CH –6, δ 7.8 and 8.1ppm (2s,2H, 2NH) , and δ9.6ppm corresponding to NH proton. ^{13}C-nmr ($CDCl_3$): δ 175.8 ppm corresponding to *C*O, δ157.4 ppm corresponding to *C* NH, δ

123.2 corresponding to cyano group, δ 38.95 CH-6, δ143.54ppm corresponding to-C-Ar, δ 127.33ppm corresponding to C-CN.

These results prompted us to continue investigation of the reactivity of substituted ethyl (2*Z*)-2-cyano-3-(substituted) phenylacrylate **6a-c** towards fatty acid hydrazide **1b** . The reaction may be proceed as in scheme1 affording 6-alkyl 3-aryl-5,7-dioxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile **7a-c**.



1 b,n=5 2 a,Ar=C_6H_2-$(OCH_3)_3$,3,4,5 b,Ar=C_6H_4-F-*p* c. Ar=C_6H_4-NO_2-*p* 7 a. n=5 Ar=C_6H_2-$(OCH_3)_3$,3,4,5 b.n=5 Ar=C_6H_4-F-*p* c,n=5 Ar=C_6H_4-NO_2-*p*

Scheme 2

The ^{13}C nmr ($CDCl_3$) spectrum of **7b** showed two signals assigned to carbon atoms of two carbonyl group at 176.9,179.142 as characteristic signals for the this structure.

In summary, we have achieved an unexpected synthesis of interesting 1,2- diazapinone derivatives via the reaction of unsaturated nitriles and fatty acid hydrazides .

EXPERIMENTAL

Melting points were taken on a Boetius melting point microscope and are uncorrected . Microanalyses were performed by Microanalytical Unit , National Research Center (Satisfactory microanalysis were obtained C ± 0.40; H ± 0.27; N ± 0.30) . IR spectra were recorded on a Mattson 5000 FT-IR Spectrophotometer . ^{1}HNMR and ^{13}CNMR spectra were determined on a JEOL Hz Spectrometer and Varian Unity Plus , using tetramethylsilane as the internal standard .Mass spectra (MS) were recorded on a Finigan SQ 700 Mass Spectrometer .

Silica gel with fluorescent indicator 254 nm on aluminum sheets layer thickness 0.2 mm were used for Thin Layer Chromatography (TLC) . Chloroform was used as eluent system for Thin Layer Chromatography .

Preparation of 1,2- Diazepinone Derivatives 5a-o and 7a-c

General method

To a solution of the fatty acid hydrazide[1] **1a-d** (0.01mole) in 20 ml ethanol, 0.02 mole of the appropriate nitrile derivative was added , and a catalytic amount of DBU. The reaction mixture was stirred at room temperature and monitored by TLC. The solid that separated was collected ,filtered off, washed with cold diethyl ether and dried affording **5 a-o** and **7 a-d**

6-butyl-5-imino-3-(3,4,5-trimethoxyphenyl)-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5a, 78%, m.p.134-135^{0}C, $C_{19}H_{24}N_4O_4$ (372.42), from diethyl ether, IR (γ/cm^{-1}) 3195 (NH), 2225 (CN), 1625 (CO) ^{1}H-NMR($CDCl_3$): 0.9 (t, 3H, CH_3), 1.43 (m, 4H, 2 x CH_2), 1.72-1.78 (m,

2H, CH_2), 2.78 (t, 1H, CH. 6), 3.9 (s,9H,9xOCH_3-3,4,5), 6.93 (s, 2H, Ar-2,6), 7.29 (s, 1H, NH exchangeable with D_2O), 7.7 (s, 1H, NH exchangeable with D_2O), 9.98 (s, 1H, NH exchangeable with D_2O), Ms:m/z(%)M^{+2}-$CH(CN)_2$, (308,100%); (209, 35%); (193,80%).

3-(2 bromophenyl)-6-butyl-5-imino-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5b, , 72%, m.p.73-74^{0}C, $C_{16}H_{17}BrN_4O$ (361.24), from pet.ether(40-60 °C), IR (γ/cm^{-1}) 3067(NH), 2220 (CN), 1665 (CO) , ^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.4 (m, 4H, 2x CH_2), 1.74-1.79 (m,2H,CH_2), 2.7(t,1H,CH-6), 7.2-7.6 (m,4H, Ar-H), 8.1 (s,1H,NH exchangeable with D_2O), 8.4 (s,1H,NH exchangeable with D_2O), 9.5 (s,1H,NH exchangeable with D_2O), ^{13}C-nmr ($CDCl_3$): δ 176.6 ppm corresponding to *C*O , δ 158.8 ppm corresponding to *C* NH, δ 124 corresponding to cyano group, δ 35.2 CH-6 , δ 32.5, 31.4, 24.8, 13.9ppm corresponding to aliphatic chain, δ 142.4, 124.11, 133.05, 130.55, 127.46, 128.23 ppm corresponding for aryl, δ 146.04ppm corresponding to-C-Ar, δ 128.23ppm corresponding to C-CN, Ms:m/z(%) M^{+2}-$CH(CN)_2$, 297 [(M^+, Br^{79}, 100%)]; 299 [(M,Br^{81}, 93%)]; 197 [(M, Br^{79}, 13%)]; 199 [(M, Br^{81}, 6%)].

6-butyl-5-imino-3-(4-nitrophenyl)-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5c, , 65%, m.p143-144^{0}C, $C_{16}H_{17}N_5O_3$ (327.34), from pet.ether(40-60), IR (γ/cm^{-1}) 3100(NH), 2230 (CN), 1690 (CO) , ^{1}H-NMR($CDCl_3$): 1.0 (t,3H,CH_3), 1.3-1.4 (m, 4H, 2x CH_2), 1.74-1.79 (m,2H,CH_2), 2.6(t,1H,CH-6), 7.2-7.8 (m,4H, Ar-H), 8.4 (s,1H,NH exchangeable with D_2O), 8.7(s,1H,NH exchangeable with D_2O), 9.8 (s,1H,NH exchangeable with D_2O).

6-butyl-5-imino-3-(4-flourophenyl)-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5d 70%, m.p122-123^{0}C, $C_{16}H_{15}FN_4O$ (298.32), from pet.ether(40-60 °C), IR (γ/cm^{-1}) 3080(NH), 2225 (CN), 1670 (CO) , ^{1}H-NMR($CDCl_3$): 1.0 (t,3H,CH_3), 1.3-1.4 (m, 4H, 2x CH_2), 1.6-1.7(m,2H,CH_2), 2.6(t,1H,CH-6), 7.2-7.6 (m,4H, Ar-H), 8.2 (s,1H,NH exchangeable with D_2O), 8.6(s,1H,NH exchangeable with D_2O), 9.4 (s,1H,NH exchangeable with D_2O).

6-hexyl-5-imino-3-(3,4,5-trimethoxyphenyl)-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5e 78%, m.p108-110^{0}C, $C_{21}H_{28}N_4O_4$ (400.47), from pet.ether(40-60 °C), IR (γ/cm^{-1}) 3187(NH), 2225 (CN), 1651 (CO) , ^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.29-1.39 (m,8H, 4xCH_2), 1.8 (m,2H,CH_2), 2.7 (t,1H,CH-6), 3.88 (s, 3H,OCH_3), 3.9 (s,6H,2xOCH_3), 6.9 (s,2H,Ar), 7.6 (s,1H,NH exchangeable with D_2O), 7.8 (s, 1H, NH exchangeable with D_2O), 10.13 (s, 1H, NH exchangeable with D_2O), M^{+2}-$CH(CN)_2$, 336 (35%);193 (100%).

7-(2-bromophenyl)-6-hexyl-5-imino-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5f 78%, m.p108-110^{0}C, $C_{21}H_{28}N_4O_4$ (400.47), from pet.ether(40-60 °C), IR (γ/cm^{-1}) 3200(NH), 2235 (CN), 1700 (CO) , ^{1}H-NMR $CDCl_3$): 0.9 (t,3H,CH_3), 1.29-1.39 (m,8H, 4xCH_2), 1.8 (m,2H,CH_2), 2.7 (t,1H,CH-6), 3.88 (s, 3H,OCH), 3.9 (s,6H,2xOCH_3), 6.9 (s,2H,Ar), 7.6 (s,1H,NH exchangeable with D_2O), 7.8 (s, 1H, NH exchangeable with D_2O), 10.13 (s, 1H, NH exchangeable with D_2O).

6-hexyl-5-imino-3-(4-nitrophenyl)-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5g 70%, m.p148-150^{0}C, $C_{18}H_{21}N_5O_3$ (355.35), from pet.ether(40-60 °C), IR (γ/cm^{-1}) 3187(NH), 2225 (CN), 1651 (CO) , ^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.20-1.30 (m,8H, 4xCH_2), 1.9 (m,2H,CH_2), 2.6 (t,1H,CH-6), 7.6-8.2 (m,5H,Ar+NH, exchangeable with D_2O), 7.5 (s,1H,NH exchangeable with D_2O), 9.7 (s, 1H, NH exchangeable with D_2O).

3-(4-fluorophenyl)-6-hexyl-5-imino-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5h 65%, m.p125-127^{0}C, $C_{18}H_{21}FN_4O$ (328.38), from pet.ether(40-60 °C), IR (γ/cm^{-1}) 3150(NH), 2230(CN), 1690 (CO) , ^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.20-1.30 (m,8H, 4xCH_2), 1.9 (m,2H,CH_2),

2.4 (t,1H,CH-6), 7.6-8.2 (m,5H,Ar+NH, exchangeable with D_2O), 7.4 (s,1H,NH exchangeable with D_2O), 9.3 (s, 1H, NH exchangeable with D_2O).

5-imino-6-octyl-7-oxo-3-(3,4,5-trimethoxypheny)l-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5k 80%, m.p136-137^{0}C $C_{23}H_{32}N_4O_4$ (428.52), from pet.ether(40-60 °C), IR (γ/cm^{-1}) 3066(NH), 2235(CN), 1662 (CO) , ^{1}H-NMR($CDCl_3$): 0.85 (t,3H,CH_3), 1.27-1.4 (m,12H, 6x CH_2), 1.7 (m,2H,CH_2),2.6(t,1H,CH-6), 3.88 (s,3H,OCH_3), 3.9 (s,6H,2xOCH_3), 6.93 (s,2H,Ar), 7.8 (s, 1H, NH exchangeable with D_2O), 8.4 (s, 1H, NH exchangeable with D_2O), 10.4 (s, 1H, NH exchangeable with D_2O), M^{+2}-$CH(CN)_2$, 364 (60%);193 (100);178 (25%).

5-imino-6-octyl-7-oxo-3-(4-nitropheny)l-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5l : 82%, m.p156-157^{0}C ,$C_{20}H_{35}N_5O_3$ (383.20), IR (γ/cm^{-1}) 3180(NH), 2226(CN), 1664 (CO) , ^{1}H-NMR($CDCl_3$): 0.8 (t,3H,CH_3), 1.3-1.4 (m,12H, 6xCH_2), 1.6-1.7 (m,2H,CH_2), 2.7 (t,1H,CH -6), 7.29 (s,1H, NH exchangeable with D_2O), 7.6-7.7 (dd,2H,Ar, J = 8.4 Hz), 7.8 (s,1H, NH exchangeable with D_2O), 8.3-8.4 (dd, 2H, Ar, J = 8.4 Hz), 9.98 (s,1H, NH exchangeable with D_2O), M^+-$CH(CN)_2$, 320 (15%);193 (100).

6-decyl-5-imino-7-oxo-3-(3,4,5-trimethoxypheny)l-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5m : 84%,m.p.112-113^{0}C, $C_{25}H_{36}N_4O_4$ (456.58), from petroleum ether (40-60 °C) IR (γ/cm^{-1}) 3190(NH), 2230(CN),1665 (CO), ^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.4-1.25 (m,14H, 7xCH_2), 1.7 (m,3H,CH_2+CH), 2.7 (t,1H,CH -6), 3.88 (s,3H,OCH_3), 3.9 (s,6H,2xOCH_3), 6.9 (s,2H,Ar), 7.8, 8,2 (2s,2H,2NH exchangeable with D_2O), 10.5 (s,1H,NH exchangeable with D_2O)., M^{+2}-$CH(CN)_2$, 392 (40%);193 (100).

6-decyl-5-imino-7-oxo-3-(4-nitrophenyl)-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5n 88%,m.p.122-123^{0}C, $C_{22}H_{29}N_5O_3$ (411.50),from petroleum ether (40-60 °C), IR (γ/cm^{-1}) 3090(NH), 2230(CN),1665 (CO), ^{1}H-NMR($CDCl_3$): 0.8 (t,3H,CH_3), 1.4 (m,16H, 8 x CH_2), 1.7 (m,2H,CH_2), 2.6 (t, 1H,CH-6), 7.6-7.7 (dd,2H,Ar, J=8.5 Hz), 7.8 (s,1H,NH exchangeable with D_2O), 8.1 (s,1H,NH exchangeable with D_2O), 8.4-8.3 (dd, 2H,Ar, J = 8.5 Hz), 9.6 (s,1H, NH exchangeable with D_2O),), ^{13}C-nmr ($CDCl_3$): δ 175.8 ppm corresponding to *C*O , δ 157.4 ppm corresponding to *C* NH, δ 123.2 corresponding to cyano group, δ 38.95 CH-6 , δ 34.47, 31.93, 31.22, 28,95, 28.84, 28.74, 24.997, 24.063,21.997, 13.515ppm corresponding to aliphatic chain, δ 147.24,126.77, 123.83, 139.64, 130.93, 123.31ppm corresponding for aryl, δ 143.54ppm corresponding to-C-Ar, δ 127.33ppm corresponding to C-CN , M^{+1}-$CH(CN)_2$, 348 (10%);193 (100).

3-(2-bromophenyl)-6-decyl-5-imino-7-oxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 5o: 85%,m.p.82-83^{0}C, $C_{22}H_{29}BrN_4O$ (445.40),from petroleum ether (40-60 °C), IR (γ/cm^{-1}) 3070(NH), 2195(CN),1665 (CO), ^{1}H-NMR($CDCl_3$): 0.84 (t,3H,CH_3), 1.2-1.6 (m, 16H,8xCH_2), 1.7 (m,2H,CH_2), 2.7 (t,1H,CH -6), 7.2-7.6 (m,4H,Ar), 7.8 (s,1H,NH exchangeable with D_2O), 8.1 (s,1H,NH exchangeable with D_2O), 10.0 (s,1H, NH exchangeable with D_2O), ^{13}C-nmr ($CDCl_3$): δ 176.5 ppm corresponding to *C*O , δ 158.7 ppm corresponding to *C* NH, δ 124.20 corresponding to cyano group, δ 35.02 CH-6 , δ 32.616, 31.826, 29.562, 29.436, 29.31, 29.224, 25.536, 24.656, 22.54, 14.054ppm corresponding to aliphatic chain, δ 146.037, 127.26, 133.989, 134.897, 130.961, 133.082 ppm corresponding for aryl, δ 142.23 ppm corresponding to-C-Ar, δ 127.56ppm corresponding to C-CN.

6-butyl-5,7-dioxo-3-(3,4,5-trimethoxyphenyl)-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 7a: 75%, m.p.77-78°C, $C_{19}H_{23}N_3O_5$ (373.40),from petroleum ether (40-60 °C), IR (γ/cm ^{1}) 3370(NH), 2190(CN),1665 (CO)

^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.2 (m, 4H,2xCH_2), 1.7 (m,2H,CH_2), 2.9 (t,1H,CH -6), 4.1(s,9H,3xOCH_3), 7.1 (s,2H,Ar-2,6), 7.8 (s,1H,NH exchangeable with D_2O), 8.1 (s,1H,NH exchangeable with D_2O).

6-butyl-3-(4-flourophenyl)-5,7-dioxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 7b : 70%, m.p. 68-69°C, $C_{16}H_{16}FN_3O_2$ (301.32),from petroleum ether (40-60 °C), IR (γ/cm^{-1}) 33550(NH), 2230(CN),1645 (CO), ^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.3 (m, 4H,2xCH_2), 1.8 (m,2H,CH_2), 3.0 (t,1H,CH -6), 7.2-7.9 (m,5H,Ar + NH, exchangeable with D_2O), 8.4 (s,1H,NH exchangeable with D_2O). ^{13}C-nmr ($CDCl_3$): δ 176.9 and 179.1 ppm corresponding to two *C* O, δ 123.20 corresponding to cyano group, δ 34.5 CH-6 , δ 31.81,29.36,29.32,29.24,29.2,29.82,22.61,14.06 ppm corresponding to aliphatic chain, δ 165.498(244HJz), 116.802(24.9 Hz),128.859(9.6Hz) 130.243 attributed for aromatic ring. 142.43(C5),136.243(C6), δ 142.43 ppm corresponding to-C-Ar, δ 136.6 ppm corresponding to C-CN.

6-butyl-3-(4-nitrophenyl)-5,7-dioxo-2,5,6,7-tetrahydro-1*H*-1,2-diazepine-4-carbonitrile 7c : 65%, m.p. 98-100°C, $C_{16}H_{16}N_4O_4$ (328.32), from petroleum ether (40-60 °C), IR (γ/cm^{-1}) 3370(NH), 2280(CN),1680 (CO), ^{1}H-NMR($CDCl_3$): 0.9 (t,3H,CH_3), 1.2 (m, 4H,2xCH_2), 1.9 (m,2H,CH_2), 3.1 (t,1H,CH -6), 7.6-8.5 (m,6H,Ar + 2NH, exchangeable with D_2O).

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