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Synthesis and characterization of the new type of tetrazoles containing barbiturates

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ABSTRACT

Reaction of 4-cyano benzaldehyde with sodium azide in presence of ammonium Chloride in dimethyl formamide under reflux conditions gave 4-(1H-tetrazole-5-yl)benzaldehyde, which was condense with barbiturates under knoevenagel condensations to give new substituted derivatives of tetrazoles with good yield.

KEYWORDS

Synthesis;
Characterization;
Tetrazole;
Barbituric acid.

INTRODUCTION

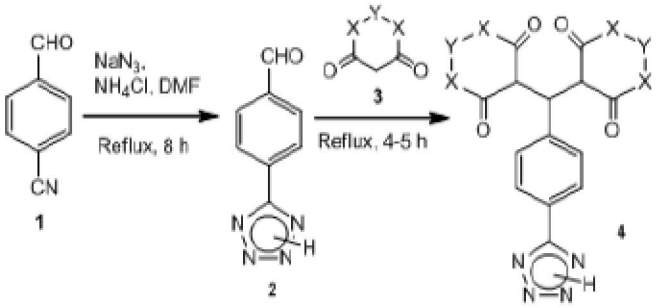
5-Substituted tetrazoles are reported to possess antibacterial[1-3], antifungal[4], antiviral[5-7], analgesic[8-12], anti-inflammatory[13-16], antiulcer[17-19] and antihypertensive[20, 21] activities. The tetrazole function is metabolically stable[22-24]. They are used as *cis*-peptide bond mimics, drugs in pharmaceuticals and bioisosteres for carboxylic acids[25]. This feature and a close similarity between the acidic character of the tetrazole group and carboxylic acid group[26] have inspired medicinal chemists to synthesize substituted tetrazoles as potential medicinal agents. Tetrazoles are an increasingly important functionality, not only as precursors to a variety of nitrogen-containing heterocycles[27] but also as materials with applications in explosives[28] and even as increasive lubricants[29]. Since the 1980s, the growth of tetrazole chemistry has continued unabated due to its popular functionality with a wide range of applications and also has been synthesized under various conditions[30]. The conventional synthesis of 5-substituted tetrazoles involves a [2+3] cycloaddition of an azide and a nitrile. But the major drawbacks of this approach are the *in situ* generation of hydrazoic acid which is highly toxic and explosive. Use of expensive and toxic metals, *etc.* an alternate methodology to overcome these shortcomings was developed for the first time by Sharpless *et al.*[31].

On the other hand, in the realm of biological chemistry, work involving artificial, hydrogen bonding receptor for barbiturate drugs[32] has inspired the preparation of barbiturate derivatives possessing specific host-guest recognition properties[33]. Barbiturate groups are strongly electron-withdrawing because they gain aromatic stabilization upon reduction[34]. This property of barbiturate has been exploited in the preparation of molecules which possess very pronounced quadratic non liner optical (NLO) properties of interest for potential applications in opto-electronic and photonic technologies[35]. The reaction of 1,3-diethyl thiobarbituric acid (DETBA) with 4-cyanobenzaldehyde by Knoevenagel condensation then subsequently Michael

addition via second molecule of DMBA were afforded 4-((bis(1,3-diethyl-4,6-dioxo-2-thioxohexahydropyrimidin-5-yl)methyl)benzonitrile[36]. This unexpected reaction outcome stimulated us to engage in synthesis of the another barbiturates such as; barbituric (BA), 1,3-dimethyl barbituric (DMBA), 1-methyl barbituric (MBA), thiobarbituric acid (TBA) and dimedone derived from 4-cyanobenzaldehyde. In this work, we interested to syntheses of 4-(1*H*-tetrazol-5-yl) benzaldehyde (2) from 4-cyanobenzaldehyde (1) then its Knoevenagel condensation and subsequently Michael addition to preparation of tetrazole containing bis barbiturates.

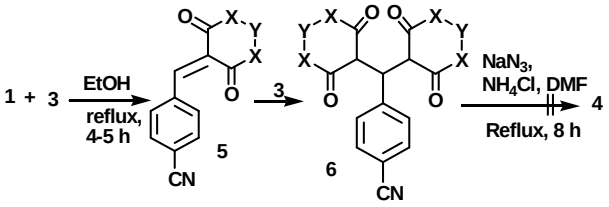
RESULTS AND DISCUSSION

This paper describes the reaction of 4-(1*H*-tetrazol-5-yl) benzaldehyde 1 with barbituric acid derivatives by Knoevenagel condensation followed by Michael addition were afforded new class of 5-substituted tetrazoles containing bisbarbiturates (Scheme 1). Initially, in the first stage, we synthesized 4-(bis(barbitur-5-yl)methyl)benzonitriles (6a-e) and dimedone derivative of 4-(bis(4,4-dimethyl-2, 6-dioxocyclohexyl) methyl)benzonitrile (6f) (Scheme 2). Our aim was attempt to conversion of nitrile group to tetrazole function via [2+3] cycloaddition reaction of sodium azide (preparation of 2). These reactions of tetrazole synthesis were filed in results because of the barbiturate ring decomposition in tetrazole working up. For this reason, first, we performed the reaction of 1 with sodium azide in the presence of catalytic amount of ammonium chloride obtained 4-(1*H*-tetrazol-5-yl) benzaldehyde 2 in good yield (Scheme 1). The reaction of 2 with β-dicarbonyl compounds such as; BA (3a), DMBA (3b), 1-methyl BA (3c), TBA (3d), DETBA (3e) and dimedone (3f) were afforded related new 5-substituted tetrazole containing bisbarbiturates (4a-e) and related dimedone derivative (4f), respectively. It is clear that compounds 5a-e results from the Michael addition of a second molecule of β-dicarbonyl compounds across vinyl group of 5. The reaction of BA 3a, DMBA 3b and 1-methyl BA 3c with 1 also typically afford Knoevenagel products (5a, 5b and 5c, respectively), the only reported exception being salicylaldehyde[36, 37]. However, no explanation was offered for the production of Michael adduct in the later case[37]. Instead, variation of the reaction stoichiometry and/or temperature affects the yield of 6a-f, but we have detected amount of 5a, 5b and 5c. For this reason, the reaction stoichiomery was carried out in the mole ratio of 1:2 for compounds 1 and 3a-f respectively.



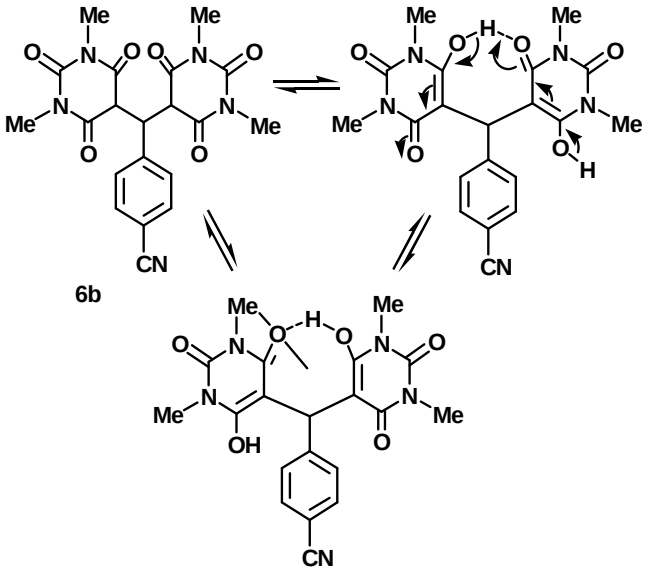
X-Y-X = NH-CO-NH (**a**), MeN-CO-NMe (**b**), MeN-CO-NH (**c**) NH-CS-NH (**d**), EtN-CS-NEt (**e**), CH_2-C(Me)$_2$-CH_2 (**f**)

Scheme 1 : Syntheses of 4-(1*H*-tetrazol-5-yl)benzaldehyde (2) from 4-cyanobenzaldehyde and Knoevenagel condensation then subsequently Michael addition with β-dicarbonyl compounds (3a-f) for preparation of 4a-f.



X-Y-X = NH-CO-NH (a), MeN-CO-NMe (b), MeN-CO-NH (c), NH-CS-NH (d) , EtN-CS-NEt (e)

Scheme 2 : Reaction of 4-cyanobenzaldehyde 1 with β-dicarbonyl compounds (3a-f).



Scheme 3 : Tautomeric forms of 6b as representative

In the [2+3] cycloaddition reaction of 1 with sodium azide for the preparation of 2 and comparison of their IR spectral data, the loss of nitrile absorption in the frequency of 2220 cm^{-1} and appearance of tetrazole NH absorption in the range of 2500-3500 cm^{-1} are the best evidences for the preparation of 2 (see experimental).

Another evidence for this conversion is ^{13}C NMR spectroscopy. ^{13}C NMR spectrum of 2 shows six distinct peaks and the peak at δ 110 ppm (in 1 related to CN) disappeared and instead a peak at δ 160 ppm was appeared in 2 (related to CN_4H carbon atom).

EXPERIMENTAL SECTION

General

Melting points were measured with a digital melting point apparatus (Electrothermal) and were uncorrected. IR spectra were determined in the region 4000-400 cm^{-1} on a NEXUS 670 FT IR spectrometer by preparing KBr pellets. The ^1H and ^{13}C NMR spectra were recorded on Bruker 300 FT-NMR at 300 and 75 MHz, respectively (Urmia University, Urmia, Iran). ^1H and ^{13}C NMR spectra were obtained on solution in $CDCl_3$ as solvents using TMS as internal standard. The data are reported as (s=singlet, d=doublet, t=triplet m=multiplet or unresolved, bs=broad singlet, coupling constant(s) in Hz, integration). All reactions were monitored by TLC with silica gel-coated plates (CCl_4: AcOEt / 80:20/ v:v). The mass analysis performed using mass spectrometer (Agilent Technology (HP) type, MS Model: 5973 network Mass selective detector Electron Impact (EI) 70 (eV), ion source temperature was 230 ºC (Tehran University, Tehran, Iran). Compounds 2a-e were synthesized in our laboratory based on reported references[38]. 4-Cyano benzaldehyde 1 and dimedone 2f and used solvents were purchased from Merck and Aldrich without further purification.

General procedure for the preparation of compounds 6a-f.

In a round bottom flask equipped with a magnetically stirrer dissolved 4-cyano benzaldehyde 1 (3.8 mmol) and barbituric acid 3a (3.8 mmol) in 10 mL ethanol and refluxed for 4-5 hrs. The white solid precipitated, filtered off then washed with cold ethanol. The crude product recrystalized from the mixture of acetone: Chloroform (1:1/v:v). (Yield 38.5%, 54.6 mg).

4-(bis(2,4,6-trioxohexahydropyrimidin-5-yl)methyl)benzonitrile (6a).

Pale White solid, mp 294°C (decomps.). IR (KBr, cm^{-1}): 3180 (–OH), 2925 (C–H, aliph.), 2231 (–CN), 1716 (C=O), 1650 (C=O), 1607, 1522 (C=C, ar.), 1289 (C–N), 1238 (C–O), 857 (para subs.); ^1H NMR (300 MHz, DMSO-d_6) δ 5.76 (bs, 2H), 7.63 (d, 4H), 7.72 (bs, 1H) 7.88 (d, *J*= 5.1Hz, 2H), 7.96 (t, 2H), 8.05 (d, *J*= 5.1Hz, 2H), 8.26 (d, *J*= 5.1Hz, 1H); ^{13}C NMR (75 MHz, DMSO-d_6) δ 193.03, 165.01, 163.19, 152.12, 150.58, 133.62, 132.38, 132.06, 131.97, 130.32, 128.42, 119.66, 108.27.

4-(bis (1,3-dymethyl-2,4,6-trioxo hexahy dropyrimidin-5-yl) methyl) benzonitrile (6b).

White crystal, mp 173 °C. IR (KBr, cm^{-1}): 3397 (–OH), 2951 (C–H, aliph.), 2531 (w), 2226 (–CN), 1704 (C=O), 1620 (C=O), 1474 (–CH_3), 1244 (C–N), 1207 (C–O), 865 (para subs.); ^1H NMR (300 MHz, $CDCl_3$) δ 3.37 (s, 6H), 3.45 (s, 6H), 5.6 (s,1H) 7.3 (d, *J*= 7.5Hz, 2H), 7.6 (d, *J*= 7.8 Hz, 2H), 10.1 (bs, 1H), 13.4 (bs, 1H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 164.7, 163.2, 150.3, 142.6, 132.2, 127.3, 118.7, 110.5, 92.6, 34.8, 29.3, 29.0.

4-(bis(1-methyl-2,4,6-trioxo hexahydro pyrimidin-5-yl) methyl) benzonitrile(6c).

Pale yellow crystal, mp 270°C (decomps.). IR(KBr, cm^{-1}): 3239 (–NH), 3109 (C–H, ar.), 2238 (–CN), 1751 (C=O), 1706 (C=O), 1658 (C=C, aro.), 1596 (–CH_3), 1291 (C–N), 849 (para subs.); ^1H NMR (300 MHz, DMSO-d_6) δ 3.07 (s, 3H), 3.17 (s, 3H), 7.93 (m, 8H), 8.06 (d, *J*= 2.4Hz, 1H), 8.31 (d, *J*= 7.8 Hz, 2H), 11.5 (s, 1H), 11.66 (s, 1H); ^{13}C NMR (75 MHz, DMSO-d_6) δ 163.01, 162.02, 160.62, 152.79, 152.28, 151.01, 150.93, 138.55, 138.41, 133.62, 132.32, 132.12, 131.97, 130.31, 128.31, 122.31, 122.24, 118.98, 113.17, 113.04, 28.14, 27.57, 27.38.

4-(bis(4,6dioxo-2-thioxohexahydropyrimidin-5-yl)methyl)benzonitrile(6d).

Yellow solid, mp 300°C (decomps.). IR (KBr, cm^{-1}): 3174 (–OH), 2924 (C–H, aliph.), 2236 (–CN), 1705 (C=O), 1672 (C=O), 1596 (–CH_3), 1377 (C–

N), 840 (para subs.); ^{1}H NMR (300 MHz, DMSO-d_6) δ 6.01 (s, 1H), 7.61 (d, *J*= 7.8 Hz, 1H), 7.61 (d, *J*= 7.8 Hz, 1H), 7.89 (d, 2H), 8.04 (d, 4H), 8.28 (d, 1H), 10.09 (d, 1H), 11.59 (bs, 3H), 11.35 (bs, 1H), 12.49 (bs, 1H); ^{13}C NMR (75 MHz, DMSO-d_6) δ 173.51, 163.4, 133.6, 132.6, 132.1, 132.0, 130.3, 128.09, 95.5.

4-(bis(1,3-diethyl-4,6dioxo-2-thioxo hexahydropyrimidin-5-yl) methyl)benzonitrile (6e). White crystal, mp 200 °C. IR(KBr, cm^{-1}): 2976 (C–H, aliph.), 2227 (–CN), 1615 (C=O), 1269 (C–N), 782 (para subs.); ^{1}H NMR (300 MHz, $CDCl_3$) δ 1.3 (t, 6H), 1.38 (t, 6H), 4.6 (m, 8H), 5.67 (s, 1H), 7.27 (d, *J*= 7.8 Hz, 2H), 7.63 (d, *J*= 8.1 Hz, 2H), 8.5 (bs, 1H), 13.9 (bs, 1H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 174.6, 163.8, 162.2, 141.6, 132.2, 127.3, 118.6, 110.8, 92.5, 45.2, 44.6, 35.3, 12.01, 11.99.

4-(bis(4,4-dimethyl-2,6-dioxo cyclohexyl) methyl)benzonitrile (6f). White crystalin solid, mp 193.5 °C. IR(KBr, cm^{-1}):3329 (–OH), 2961 (C–H, aliph.), 2226 (–CN), 1721 (C=O), 1662 (C=O), 1471 (–CH_3), 847 (para subs.); ^{1}H NMR (300 MHz, $CDCl_3$) δ 0.99 (s, 6H), 1.12 (s, 6H), 1.54 (bs, 1H), 2.21 (q, 4H), 2.5 (s, 4H), 4.78 (s, 1H), 7.42 (d, *J*= 7.8Hz, 2H), 7.53 (d, *J*= 8.1 Hz, 2H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 196.17, 162.8, 149.4, 131.9, 129.2, 114.6, 110.2, 50.6, 40.8, 32.4, 32.2, 29.2, 27.2.

General procedure for the preparation of compounds 2

In a round bottom flask equipped with a magnetically stirrer dissolved 4-cyano benzaldehyde 1 (0.25 gr, 2 mmol), NH_4Cl (0.102 gr, 2 mmol) and sodium azide (0.12 gr, 2 mmol) in 3 mL DMF and refluxed for 8 hrs. The solution of reaction mixture acidified to pH = 2 in an ice-bath. Added 100 mL water and 4-(1*H*-tetrazol-5- yl)benzaldehyde (2) was precipitated as a white solid (Yield 56%, 185 mg).

General procedure for the preparation of compounds 4a-f

In a round bottom flask equipped with a magnetically stirrer dissolved 4-(1*H*-tetrazol-5-yl)benzaldehyde 2 (3.8 mmol) and barbituric acid 3b (3.8 mmol) in 10 mL ethanol and refluxed for 4-5 hrs. The white solid precipitated, filtered off then washed with cold ethanol. The crude product recrystalized in ethanol. (Yield 73.7%, 59 mg).

5,5'-((4-(1*H*-tetrazol-5-yl) phenyl) methylene) bis(1,3-dimethylpyrimidine-2,4,6(1*H*,3*H*,5*H*)-trione) (4b).

Pale White solid, mp 268.5 °C. IR (KBr, cm^{-1}): 3438 (–OH), 2850-3089 (N–H, tet.), 1615.07 (C=O), 1495 (–CH_3), 1066 (C–N), 850 (para subs.); ^{1}H NMR (300 MHz, $CDCl_3$) δ 2.48 (s, 8H), 5.55 (s, 1H), 7.77 (d, *J*= 7.8Hz, 2H), 8.2(d, *J*= 7.8 Hz, 2H), 8.4 (bs, 2H), 11.6 (bs, 1H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 164.2, 141.5, 130.7, 128.8, 128.2, 128.1, 127.8, 124.5, 68.9.

5,5'-((4-(1*H*-tetrazol-5-yl) phenyl) methylene) bis(1-methylpyrimidine-2,4,6(1*H*,3*H*,5*H*)-trione) (4c).

Pale yellow solid, mp in 280°C (decamps.). IR (KBr, cm^{-1}): 3434.59 (–NH), 2750-3091 (N–H, tet.), 1616 (C=O), 1495 (–CH_3), 851 (para subs.); ^{1}H NMR (300 MHz, DMSO-d_6) δ 2.48 (s, 5H), 5.54 (s, 1H), 7.77 (d, 2H), 8.21 (m, 3H), 8.41 (d, *J*= 8.1 Hz, 2H), 11.7 (s, 1H);^{13}C NMR (75 MHz, DMSO-d_6) δ 136.94, 126.15, 125.54, 123.93, 123.13, 118.16, 117.42, 115.2, 113.61, 106.57, 55.75, 28.17, 27.4.

5,5'-((4-(1*H*-tetrazol-5-yl) phenyl) methylene) bis(2-thioxo dihydropyrimidine-4,6(1*H*,5*H*)-dione) (4d).

Yellow solid, mp 279°C. IR(KBr, cm^{-1}): 3433.31 (–OH), 3087.92 (N–H, tet.), 2962.28(N–H, tet.), 1615 (C=O), 852 (para subs.); ^{1}H NMR (300 MHz, DMSO-d_6) δ 5.53 (s, 1H), 7.75 (d, 3H) 8.17 (m, 3H), 8.38 (bs, 3H), 11.68 (bs, 1H); ^{13}C NMR (75 MHz, DMSO-d_6) δ 164.2, 141.5, 130.7, 129.4, 128.8, 128.2, 128.09, 127.8, 127.3, 125.4, 124.5, 68.9.

5,5'-((4-(1*H*-tetrazol-5-yl) phenyl) methylene) bis(1,3-diethyl-2-thioxo dihydropyrimidine-4,6(1*H*,5*H*)-dione) (4e).

White solid mp in 290°C (decomps.). IR (KBr, cm^{-1}): 3429 (–OH),2850-3089 (N-H, tet), 1615 (C=O), 851 (para subs.); ^{1}H NMR (300 MHz, $CDCl_3$) δ 5.54 (s, 1H), 7.76 (d, *J*= 7.5 Hz, 2H), 8.14 (d, *J*= 6.3 Hz, 3H), 8.3(bs, 2H), 11.65(bs, 1H); ^{13}C NMR (75 MHz, $CDCl_3$) δ

164.2, 141.5, 130.6, 129.2, 128.8, 128.2, 128.09, 127.7, 126.9, 124.5, 68.9. 2,2'-((4-(1*H*-tetrazol-5-yl) phenyl) methylene) bis(5,5-dimethylcyclohexane-1,3-dione) (4f).

White solid, mp 287 °C. IR (KBr, cm^{-1}): 3422 (–OH), 2850-3083 (N-H, tet), 1609 (C=O), 1495 (–CH_3), 853 (para subs.); ^{1}H NMR (300 MHz, $CDCl_3$) δ 3.6 (bs, 15H), 5.53 (s, 1H), 7.76 (d, *J*= 7.8Hz, 2H), 8.18 (d, *J*= 8.1 Hz, 2H), 8.36 (bs, 2H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 164.4, 164.2, 156.3, 155.6, 144.02, 141.5, 141.4, 133.5, 130.6, 128.8, 128.2, 127.9, 125.4, 124.1, 118.9, 112.2, 68.9, 68.7.

CONCLUSION

The method described in this paper, allows the preparation of unique substituted Tetrazoles from commerical and available nitriles and easy to preparefrom sodium azides. The important aspects of this protocol, are mild reaction conditions, availability of the percursors and purity of the obtained products with no further crystalization.

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