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Reaction of 4-*n*-propyl-1,2,4-triazoline-3,5-dione with some selected dienes

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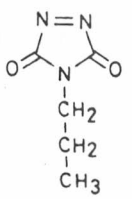
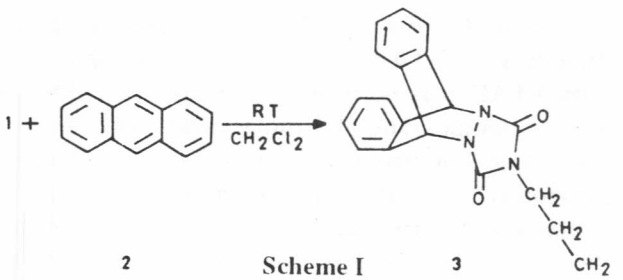
The reaction of 4-*n*-propyl-1,2,4-triazoline-3,5-dione (PTAD) **1** with anthracene, cyclopentadiene (monomer), 5,5-dimethoxy-1,2,3,4-tetrachlorocyclopentadiene, hexachlorocyclopentadiene, isoprene, 2,6-dichlorostyrene, 1,3-cyclohexadiene, and *cis, cis*-1,3-cycloheptadiene has been investigated. These reactions occur via [4+2] Diels-Alder cycloaddition with the formation of novel heterocycles in quantitative yields. The initial reaction of PTAD with 2,6-dichlorostyrene is a [4+2] Diels-Alder cycloaddition which is followed by an allylic type migration of chloride atom.

4-Substituted-1,2,4-triazoline-3,5-diones are the most reactive dienophiles[1-12], enophiles[6-12] and electrophiles[13-15]. In a previous paper[16] we have reported a convenient method for the synthesis of 4-*n*-propyl-1,2,4-triazoline-3,5-dione (PTAD) **1** in a high yield and purity. The purpose of this investigation was to examine the reactivity of PTAD with some selected dienes.



PTAD 1

The reaction of PTAD **1** with anthracene **2** was performed in methylene chloride at room temperature. Addition of the pink crystals of PTAD to a methylene chloride solution of anthracene **2** produced a purple colour which after a few min. changed to a light-pink colour and then to a pale yellow after about 60 min. At the end of reaction, the solvent was removed giving a white solid which on recrystallization from toluene-hexane afforded white needles of **3** (Scheme I). Elemental analysis showed **3** to be a 1:1 adduct formed by a [4+2] Diels-Alder cycloaddition reaction.

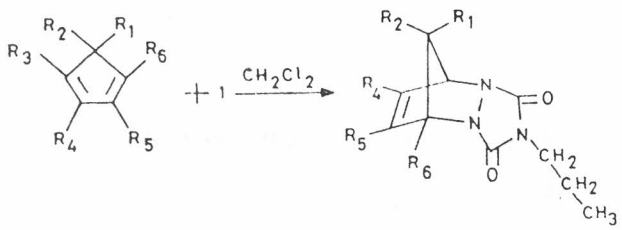


Scheme I

The ^{1}H NMR spectrum of **3** showed signals at δ 0.05 (t, methyl protons), 1.35 (sextet, methylene protons attached to the methyl group), 3.22 (t, methylene protons attached to the nitrogen atom), 6.25 (s, bridged C-H protons) and 7.2-7.6 (m, aromatic protons).

The monomeric cyclopentadiene **4** underwent reaction with PTAD (Scheme II) when a methylene chloride solution of PTAD was added dropwise to a methylene chloride solution of **4** at -10°C in 1:1 molar ratio. The reaction was extremely fast even at -10°C. The product **5** was obtained as white crystals after purification via recrystallization. The yield was quantitative and the elemental analysis agreed with 1:1 adduct.

The ^{1}H NMR spectrum of **5** showed signals at δ 0.83, 1.58 and 3.33 due to the protons of *n*-propyl group. The spectrum also exhibited peaks at δ 1.86 (d, one of the protons of the bridgehead CH_2), 2.18 (d, the other proton of the bridgehead CH_2), 5.00 (s, bridged protons) and 6.33 (s, olefinic protons).



4: $R_1=R_2=R_3=R_4=R_5=R_6=H$
5:$R_1=R_2=R_3=R_4=R_5=R_6=H$
6:$R_1=R_2=OCH_3$ $R_3=R_4=R_5=R_6=Cl$
7:$R_1=R_2=OCH_3$ $R_3=R_4=R_5=R_6=Cl$
8:$R_1=R_2=R_3=R_4=R_5=R_6=Cl$
9:$R_1=R_2=R_3=R_4=R_5=R_6=Cl$

Scheme II

The ^{13}C NMR (20 MHz) spectrum of **5** showed peaks at δ 10.50 (C'_a,) 26.71 (C_b), 40.77 (C_c), 48.13 (C_d), 64.19 (C_e), 130.53 (C_f) and 159.77 (C_g). The downfield shift in the signals of C_c and C_e carbons is due to β-effect. Similarly the reaction of 5,5-dimethoxy-1,2,3,4-tetrachlorocyclopentadiene **6** with PTAD was performed in methylene chloride at room temperature (1:1 molar ratio). The reaction was slow compared to that of cyclopentadiene (monomer) even at room temperature. This can be explained in terms of frontier molecular orbitals theory. Replacement of four hydrogen atoms by four chlorine atoms in cyclopentadiene ring, lowers the energy level of HOMO in diene **6** and therefore the energy gap between HOMO of diene **6** and LUMO of dienophile **1** increases causing a decrease in reaction rate.

The yield was quantitative and the elemental analysis agreed with 1:1 adduct **7**.

The ^1H NMR spectrum of **7** showed signals at δ 0.88, 1.60 and 3.43 which were assigned to the protons of *n*-propyl group. The signals at δ 3.6 and 3.8 (each s) were assigned to the protons of methoxy groups.

The reaction of hexachloro cyclopentadiene **8** with PTAD was also carried out in a similar way in methylene chloride at room temperature (Scheme II). The Diels-Alder cycloaddition in this case was very slow compared to that of cyclopentadiene and it required almost 48 hr for completion. In this case, two more chlorine atoms further lowered the energy level of HOMO in diene **6** leading to further large energy differences between HOMO and LUMO, and thus, the rate of reaction decreased drastically.

The yield of the cycloadduct was quantitative and the elemental analysis agreed with 1:1 adduct **9**.

The ^1H NMR spectrum of **9** showed signals at δ 0.93, 1.75 and 3.48 which were assigned to the protons of *n*-propyl group.

Isoprene **10** underwent reaction with PTAD (Scheme III) when a methylene chloride solution of PTAD was added dropwise to a methylene chloride solution of **10**. The reaction was extremely fast even at -10°C. The cycloadduct was purified from *n*-hexane. The elemental analysis was in agreement with 1:1 adduct **11**.

The ^1H NMR spectrum of **11** showed signals at δ 0.95, 1.75 and 3.65 (each s, *n*-propyl group protons), 1.86 (s, methyl protons), 4.0 (s, methylene protons), and 4.63 (br s, olefinic protons).

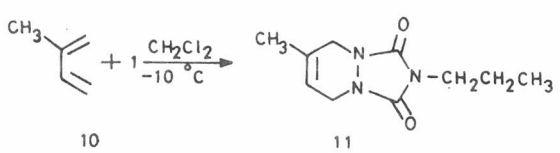
The reaction of 2,6-dichlorostyrene **12** with PTAD was also performed in methylene chloride at room temperature (Scheme IV). The product was obtained as white crystals after recrystallization from *n*-hexane.

The elemental analysis was in agreement with 1:1 adduct **14**. The first step of the reaction is the formation of a reactive intermediate **13** via a [4+2] Diels-Alder reaction. The second step is an allylic type rearrangement of chlorine atom which rearomatizes the benzene ring. Although this reactive intermediate **13** can undergo a second Diels-Alder reaction, but a double Diels-Alder adduct was not isolated.

Earlier[11] we have investigated the reaction of 4-methyl- and 4-phenyl-triazolinediones with 2,6-dichlorostyrene and the structure of resultant adduct was determined by X-ray diffraction analysis of a single crystal which confirmed the migration of chlorine atom.

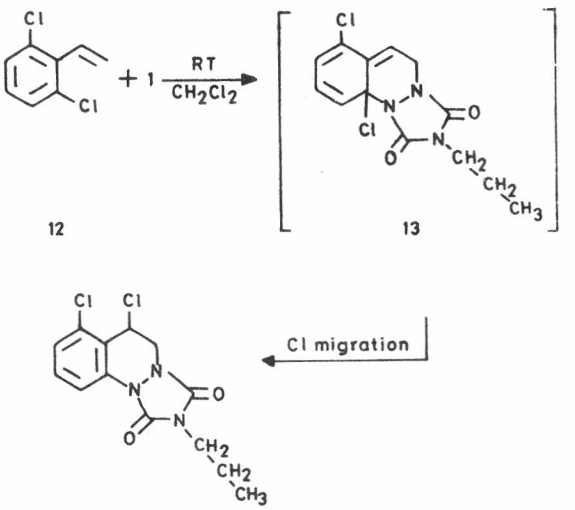
The ^1H NMR of compound **13** showed signals which are in agreement with its structure.

The reaction of PTAD with 1,3-cyclohexadiene **15** was performed in methylene chloride at -10°C.

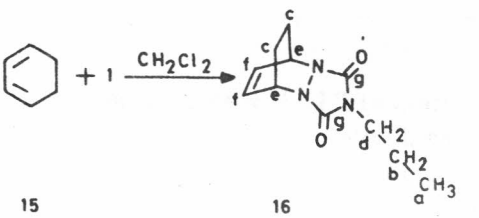


10 11

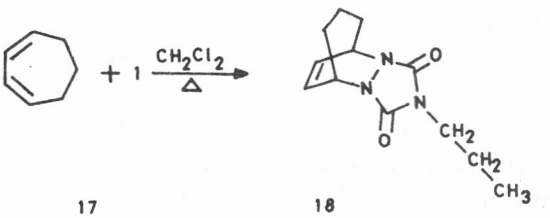
Scheme III



Scheme IV



Scheme V



Scheme VI

The reaction was extremely fast even at lower than -10°C. The completion of the reaction could be easily determined by the disappearance of the pink colour of PTAD. The yield of the cycloadduct was quantitative and elemental analysis was in agreement with 1:1 adduct **16** (Scheme V).

The ^{1}H NMR spectrum of **16** showed signals which were in agreement with the assigned structure **16**.

The ^{13}C NMR (20 MHz) spectrum of **16** showed signals at δ 10.61 (C_a), 20.88 (C_b), 21.73 (C_c), 40.55 (C_d), 49.88 (C_e), 129.86 (C_f) and 157.79 (C_g). The upfield shift in the signals of C_c and C_e carbons compared to the corresponding signals in compound **5** is due to β-effect.

The reaction of PTAD with *cis,cis*-1,3-cycloheptadiene **17** was performed in methylene chloride at refluxing temperature. The reaction was very slow and it took a long time for the pink colour of PTAD to disappear. From 1,3-cyclohexadiene to *cis,cis*-1,3-cycloheptadiene there is a drastic decrease in the reactivity of dienes. This could be attributed to the flexibility of compound **17** which does not have rigid *cis*-configuration, which is required for [4+2] Diels-Alder reaction. Besides **18** it is possible to have another product via ene reaction which was not isolated (Scheme VI).

The ^{1}H NMR spectrum of **18** showed signals which were in agreement with the assigned structure **18**.

Although PTAD is very reactive dienophile it does not react with tetramethylbenzene. The reaction of PTAD with 1,2,4,5-tetramethylbenzene was attempted in methylene chloride at refluxing temperature for 48 hr, but no reaction was observed. Tetramethylbenzene has been reported to react with benzyne intermediate to give a cycloadduct **17** in 41% yield[17].

Conclusion

It is very convenient to synthesize 4-*n*-propyl-1,2,3-triazoline-2,5-dione in a large quantity with high yield and high purity. This compound is extremely reactive dienophile. Its reaction with a wide variety of dienes gave novel heterocyclic compounds in quantitative yields. The resulted cycloadducts are very interesting compounds for further studies.

Experimental Section

General. IR spectra were recorded on a Shimadzu 435 IR spectrophotometer using KBr pellets. Vibrational transition frequencies are reported in wave numbers (cm^{-1}). Proton nuclear magnetic resonance (^{1}H NMR) spectra were recorded at 90 MHz on a Varian EM-390 instrument. Melting point were taken on a GallenKamp melting point apparatus and are incorrected. Elemental analyses were performed at the Research Institute of Petroleum Industry, Tehran, I.R. Iran.

Reagents were purchased from Aldrich Chemical Co., Fluka Chemical Co., or Riedel-deHaen AG. Isoprene was donated by Professor A

Entezami, Department of Organic Chemistry, Faculty of Chemistry, Tabriz University, IR Iran.

Reaction of PTAD with anthracene. Into a 100 mL round-bottomed flask were placed 0.62g (3.5×10^{-3} mole) of anthracene and 40 mL of methylene chloride. The flask was fitted with a magnetic stirrer. The stirrer was started and 0.50 g (3.5×10^{-3} mole) of PTAD crystals was added when a deep-purple colour appeared which later on turned to a light-pink colour. After about 60 min., a pale yellow solution was obtained. The solution was stirred at room temperature for 4hr. The solvent was evaporated under vacuum to give a white solid, yield 1.12g (100%). Recrystallization from toluene-hexane gave white needles, m.p. 179-80°; IR (KBr): 2950 (m), 2850 (w), 1770 (m), 1710 (s,br), 1570 (m,br), 1440 (s), 1410 (s), 1380 (w), 1345 (w), 1315 (w), 1165 (s), 1090 (w), 1070 (m), 1050 (m), 1030 (m), 910 (m,sh), 820 (m,sh), 800 (w), 790 (m), 765 (s), 690 (m), 535 (m) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 0.50 (t, 3H, *J*=7.5 Hz), 1.35 (s, 2H), 3.22 (t, 2H, *J*=6.0 Hz), 6.25 (s, 2H), 7.2-7.6 (m, 8H); Anal. Calcd for $C_{19}H_{17}N_3O_2$:C, 71.45; H, 5.73; N, 13.16. Found: C, 71.60; H, 5.40; N, 13.10%.

Reaction of PTAD with cyclopentadiene. Freshly distilled cyclopentadiene (0.46 g, 7.0×10^{-3} mole) and 10 mL of methylene chloride were placed into a 50mL round-bottomed flask. The solution was cooled to -10° with an ice-salt bath while being magnetically stirred. To this was added a solution of PTAD (1.0 g, 7.0×10^{-3} mole) in 10 mL of methylene chloride all at once when a pale yellow solution was obtained. Methylene chloride was removed under vacuum to leave 1.46 g (100%) of pale yellow solid which on recrystallization from hexane fraction gave white crystals, m.p. 49°; IR (KBr): 3050 (w), 3020 (m), 2950 (m), 2860 (w), 1770 (s), 1710 (s,br), 1590 (w), 1440 (s), 1410 (s,br), 1370 (m), 1330 (m), 1280 (w), 1185 (s), 1060 (s), 995 (m), 960 (m), 920 (s), 870 (m), 800 (m,sh), 780 (s), 740 (s), 660 (w), 610 (m) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 0.83 (t, 3H, *J*=9.0 Hz), 1.58 (s, 2H), 1.86 (d, 1H, *J*=9.0 Hz), 2.18 (d, 1H, *J*=9.0 Hz), 3.33 (t, 2H, *J*=7.5 Hz), 5.00 (s, 2H), 6.33 (s, 2H); Anal. Calcd. For $C_{10}H_{13}N_3O_2$: C, 57.96; H, 6.32; N, 20.28. Found: C, 57.90; H, 6.40; N, 20.20%.

Reaction of PTAD with 5,5-dimethoxy-1,2,3,4-tetrachlorocyclopentadiene. In a 50 mL round-bottomed flask, equipped with a magnetic stirrer and a constant-pressure dropping funnel, a solution of 1.85 g (7.0×10^{-3} mole) of 5,5-dimethoxy-1,2,3,4-tetrachlorocyclopentiadiene in 10 mL of methylene chloride was prepared. To this solution under stirring a pink solution of 1.00 g (7.0×10^{-3} mole) of PTAD in 10 mL of methylene chloride was added dropwise over a period of 2.5 hr. The solution developed a deep-purple colour which turned to pale yellow. The solvent was removed under vacuum to give 2.85 g (100%) of white solid which on recrystallization from methanol gave white crystals, m.p. 133°; IR (KBr): 3500 (w), 3020 (w,sh), 3000 (w,sh), 2950 (m), 2820 (w), 1795 (m), 1750 (s,br), 1580 (s), 1445 (s), 1400 (s), 1345 (m), 1210 (s), 1180 (s), 1150 (s), 1130 (s), 1100 (m), 1070 (m), 1010 (s), 980 (s), 930 (m), 890 (m), 805 (s), 780 (w), 760 (m), 650 (w), 540 (m) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 0.88 (t, 3H, *J*=9.0 Hz), 1.60 (s, 2H), 3.43 (t, 2H, *J*=7.5 Hz), 3.6 (s, 3H), 3.8 (s, 3H); Anal. Calcd for $C_{12}H_{13}N_3O_4Cl_4$: C, 35.58; H, 3.23; N, 10.37. Found: C, 35.50; H, 3.30; N, 10.20%.

Reaction of PTAD with hexachlorocyclopentadiene. Hexachlorocyclopentadiene (1.94 g, 7.0×10^{-3} mole) and 10 mL of methylene chloride were placed in a 50 mL round-bottomed flask. Stirring was started and a solution of 1.0 g (7.0×10^{-3} mole) of PTADS in 10 mL of methylene chloride was added all at once. The solution became deep-purple which turned to pink. After 48 hr a pale-yellow solution was obtained. Methylene chloride was removed under vacuum to leave 2.94 g (100%) of white solid which on recrystallization from *n*-hexane gave white needles, m.p. 103-4°; IR (KBr): 3500 (w), 2995 (m), 2990 (m), 2850 (m), 1810 (s), 1745 (s,br), 1580 (s), 1440 (s), 1400 (s,br), 1360 (s), 1340 (s), 1170 (s), 1130 (s), 1100 (s,br), 980 (w), 930 (m), 910 (m), 880 (s), 780 (s), 760 (m,sh), 720 (s), 690 (m), 630 (w), 530 (m) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 0.93 (t, 3H, *J*=7.5 Hz), 1.57 (s, 2H), 3.48 (t,2H, *J*=6.0 Hz); Anal. Calcd for $C_{12}H_7N_3O_2Cl_6$: C, 29.02; H, 1.70; N, 10.15. Found: C, 28.50; H, 1.70; N, 10.30%.

Reaction of PTAD with isoprene. Into a 100 mL two-necked round-bottomed flask, equipped with a magnetic stirrer and a thermometer, 0.72 g (1.13×10^{-3} mole) of freshly distilled isoprene and 30mL of methylene chloride were added. The solution was stirred and cooled to -10° with an ice-

salt bath. A solution of 1.6 g (1.13×10^{-3} mole) of PTAD in 25mL of methylene chloride was added dropwise over a period of 30 min. The pink colour of PTAD disappeared as fast as the solution was added to give a pale-yellow solution. The solvent was removed under vacuum to give a pale-yellow gummy solid which on recrystallization from *n*-hexane gave white crystals; 1.80 g (76%), m.p. 34-6°; IR (KBr): 3050 (w), 2950 (s), 2930 (s), 2900 (m), 2880 (m), 1765 (s), 1700 (s,br), 1560 (w), 1440 (s,br), 1415 (s), 1390 (m), 1350 (m), 1310 (w), 1260 (m,br), 1175 (w), 1070 (m), 1030 (m), 940 (w), 890 (w), 765 (m,br), 730 (w) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 0.95 (t, 3H, *J*=9.0 Hz), 1.75 (s, 2H), 1.86 (s, 3H), 3.65 (t, 2H, *J*=9.0 Hz), 4.0 (m, 4H), 4.63 (s, 1H); Anal. Calcd for $C_{10}H_{15}N_3O_2$: C, 57.40; H, 7.23; N, 20.10. Found: C, 57.30; H, 7.30; N, 20.20%.

Reaction of PTAD with 2,6-dichlorostyrene. Into a 50 mL round-bottomed flask were placed 0.60 g (3.5×10^{-3} mole) of 2,6-dichlorostyrene and 10 mL methylene chloride. The stirring was started and a solution of 0.50 g (3.5×10^{-3} mole) of PTAD in 20 mL of methylene chloride was added all at once when deep-purple colour appeared. After 9 hr a pale-yellow solution was obtained. The solvent was removed under vacuum to leave a gummy solid which was triturated with ether to yield 1.1 g (100%). Recrystallization from *n*-hexane gave white crystals, m.p. 105°; IR (KBr): 3450 (w), 3090 (w), 2950 (m), 2880 (m), 1770 (s,br), 1590 (s), 1570 (s), 1460 (s,br), 1420 (s), 1380 (s), 1350 (s), 1300 (m), 1270 (m), 1180 (s), 1110 (m), 1065 (m), 1005 (m), 950 (w), 920 (m), 780 (s), 750 (s), 705 (w), 690 (w), 620 (m,br), 500 (w) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 1.03 (t, 3H, *J*=7.5 Hz), 1.76 (s, 2H), 3.66 (t, 2H, *J*=7.5 Hz), 3.84 (dd, 1H, J_1=11.7 Hz, J_2=2.1 Hz), 4.70 (dd, 1H, J_1=11.7 Hz, J_2=2.1 Hz), 5.66 (t, 1H, *J*=2.1 Hz), 7.3 (dd, 1H, J_1=8.4 Hz, J_2=unresolved), 7.49 (t, 1H, *J*=8.4 Hz), 8.33 (dd, J_1 =8.4 Hz, J_2=unresolved); Anal. Calcd for $C_{13}H_{13}N_3O_2Cl_2$:C, 49.70; H, 4.17; N, 13.38. Found: C, 49.30; H, 4.30; N, 13.50%.

Reaction of PTAD with 1,3-cyclohexadiene. Into a 25 mL one necked round-bottomed flask, equipped with a magnetic stirrer, 0.187 g (0.22 mL, 2.3×10^{-3} mole) of 1,3-cyclohexadiene in 5 mL of methylene chloride was added. The solution was stirred by a magnetic stirrer and cooled to -10° with an ice-salt bath. A solution of 0.33 g (2.3×10^{-3} mole) of PTAD in 10 mL of methylene chloride was added dropwise. The pinck colour of PTAD disappeared as fast as the solution was added. A pale-yellow solution was obtained. The solvent was removed to give a pale-yellow gummy solid, yield, 0.517 g (100%). The gummy solid was sublimed under vacuum at 60° to give a white solid, which on recrystallization from hexane fraction gave white needles, m.p. 65°; IR (KBr): 3400 (w,br), 2950 (m), 2850 (w), 1700 (s,br), 1440 (s), 1418 (s), 1370 (m), 1325 (w), 1235 (w), 1200 (m), 1170 (m), 1110 (w), 1060 (m), 1050 (m), 950 (w), 920 (m), 890 (w), 870 (w), 850 (m), 820 (w), 760 (s), 730 (m), 690 (m), 640 (m), 580 (m) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 0.86 (t, 3H, *J*=7.5 Hz), 1.33-1.96 (m, 4H, overlapped by doublet), 2.16 (d, 2H, *J*=9.0 Hz), 3.46 (t, 2H, *J*=9.0 Hz), 4.83 (dd, 2H, *J*=unresolved), 6.46 (dd, 2H, *J*=3 Hz); Anal Calcd for $C_{11}H_{15}N_3O_2$: C, 59.70; H, 6.80; N, 19.0. Found: C, 59.90; H, 6.90; N, 19.40%.

Reaction of PTAD with *cis,-cis*-1,3-cycloheptadiene. Freshly distilled *cis,cis*-1,3-cycloheptadiene (0.76 g, 0.87 mL, 7.0×10^{-3} mole) and 10 mL of methylene chloride were placed in a 100 mL round-bottomed flask. The solution was stirred by a magnetic stirrer and a solution of 1.0 g (7.0×10^{-3} mole) of PTAD in 40 mL of methylene chloride was added all once. The solution was stirred at room temperature for 24 hr, and then refluxed for 12 hr. Solvent was removed and the resultant oil was triturated with ether and the residue sublimed under vacuum at 90° to give 0.60 g (34%) of white crystals, m.p. 107°; IR (KBr): 3300 (w), 2900 (s), 2850 (m,sh), 1740 (s), 1680 (s,br), 1450 (s), 1420 (s), 1370 (m), 1290 (m,br), 1230 (w), 1150 (w), 1050 (m), 970 (m), 890 (w), 850 (w), 810 (m), 770 (m), 750 (m), 680 (w), 640 (w) cm^{-1}; ^1H NMR ($CDCl_3$, TMS): δ 0.96 (t, 3H, *J*=7.5 Hz), 1.2-2.43 (m, 8H), 3.56 (t, 2H, *J*=7.5 Hz), 4.93 (s, br, 2H), 6.0-6.3 (m, 2H); Anal. Calcd for $C_{12}H_{17}N_3O_2$: C, 61.3; H, 7.3; N, 17.9. Found: C, 62.2; H, 7.5; N, 16.50%.

Acknowledgement

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