ESTERS AND URETHANES WITH PYRIMIDINE RING

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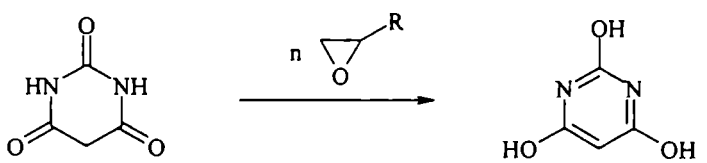
Abstract: Carboxylic acids and isocyanates react with hydroxyalkyl derivatives of 5,5-diethylbarbituric acid which were obtained from barbituric acid and oxiranes or alkylene carbonates to give corresponding esters and urethanes with pyrimidine ring. The products were isolated and identified on the basis of elemental analysis, IR and ^{1}H -NMR spectroscopy and other regular analytical methods.

Introduction

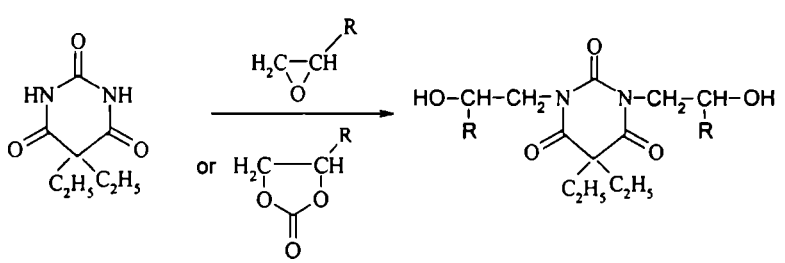
N-hydroxyalkyl derivatives of azacycles with nitrogen-bonded hydrogen are usually obtained by oxyalkylation with ethylene or propylene oxides [1-7], and more rarely with alkylene carbonates [8, 9]. Such derivatives were obtained from isocyanuric acid [1], parabanic acid [7], melamine [2], 5,5-dialkylhydantoine, 5,6-dihydrouracil [5, 6] and other substrates. The esterification of such derivatives with acrylic and methacrylic acid was also reported [10,11] leading to monomers and crosslinking agents for polymers. Nevertheless, the data on esters and urethanes with pyrimidine rings (from barbituric acid) are missing. The lack of data on those are consistent with the fact, that during the heating of barbituric acid (**1**) with oxiranes or alkylene carbonates the barbituric acid rearranges into tris-enol form (**2**), which is insoluble in those conditions and does not react with hydroxyalkylating reagents [12]:



(1) **(2)**

where R = -H, -CH_3

However, when hydrogens at C_5 are replaced with alkyl groups like in 5,5-diethylbarbituric acid (**3**) the tautomerization is not possible and renders the **3** suitable substrate for synthesis of N-hydroxyalkyl derivatives (**4**) [13]:



(3) **(4)**

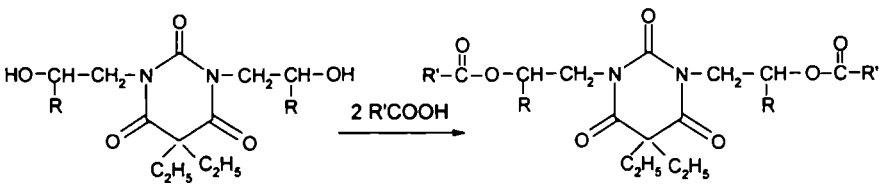
where: R = H, CH_3

This strategy enables the synthesis of new derivatives; esters and urethanes with pyrimidine ring starting from 1,3-bis(hydroxyalkyl) derivatives of 5,5-diethylbarbituric acid (BHADEB, **4**) with carboxylic acids and isocyanates, respectively. This synthetic work is presented here.

Results and Discussion

Esters

Previously obtained diols [13] were used for synthesis of esters with pyrimidine ring, including esters obtained from unsaturated acids according to the scheme:



(5)

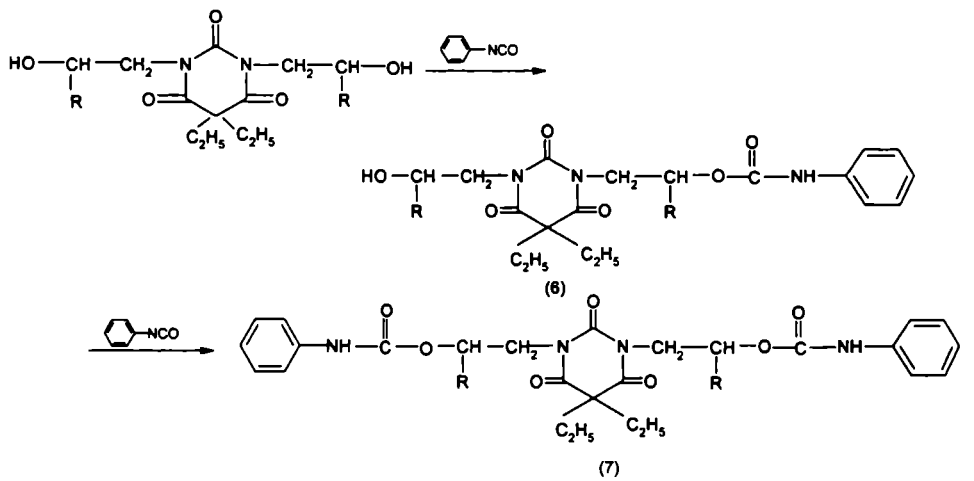
where: R = H, CH_3 ; R' = CH_3, Ph, $C_{17}H_{35}$, CH_2=CH-, CH_2=C(CH_3) -

The synthetic procedure was preliminarily established for esterification of 1,3-bis(2-hydroxyethyl) 5,5-diethylbarbiturate (BHEDEB) with acetic acid. The reaction was performed in toluene, in which the substrates were soluble and the solvent was used for aseotropic removal of water. When stoichiometric 1:2 amounts of BHEDEB and acetic acid were used, some of unreacted BHEDEB was left unreacted as identified by IR (the band 3000 cm^{-1} of ν(O-H)) and ^{1}H NMR spectral monitoring. Therefore slight excess of the acid related to BHEDEB (1.3 moles/mole of OH groups) was necessary to increase the yield of ester. Similar results were obtained with benzoic, stearic, acrylic and methacryclic acid. The reaction mixtures were neutralized with aqueous K_2CO_3, the toluene was removed on rotary evaporator. The crude ester products were purified by high-vacuum distillation. 2,2'-Bis(5,5-diethyl-2,4,6-trioxopyrimidyl)diethyl distearate was isolated by precipitation of unreacted stearic acid followed by extraction of the unreacted BHEDEB with water instead of water solution of potassium carbonate., which could not be used due to formation of soaps resulting in difficulties of phase separation. The products, except stearates are resin-like substances.

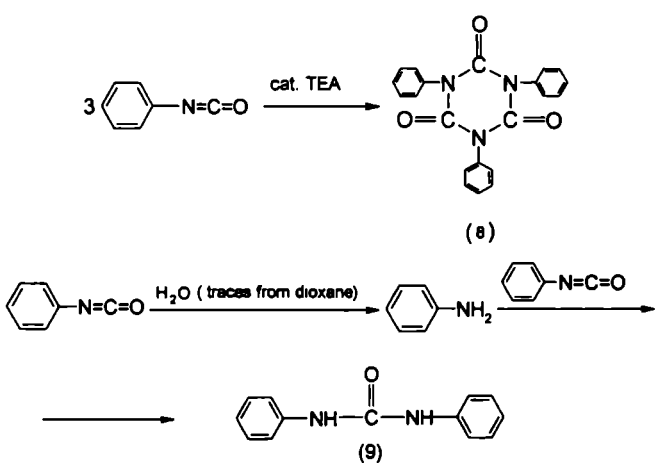
The optimized catalytic amount of sulfuric acid was 20 cm^3/mole of 1,3-bis(2-hydroxyalkyl) 5,5-diethylbarbiturate (BHADEB). In such a case the reaction was completed within 2.5 hours. The products were identified by elemental analysis and ^{1}H-NMR and IR spectra (see Experimental). In the IR spectra of products the carbonyl stretchings were observed at 1714-1745 cm^{-1} i 1670-1690 cm^{-1} regions in esters and pyrimidine ring, respectively, and the band attributed at CO-O group at 1222-1298 cm^{-1}. In the ^{1}H NMR spectra the resonances assigned to methyl and methylene protons of pyrimidine attached ethyl groups were observed at 0.7-0.8 ppm and 1.8-1.95, respectively as well as methylene or methine groups of protons (in products obtained from BHPDEB) linked with ester group (CH_2OCO or CHOCO) at 4.2-4.4 or 5.1-5.3 ppm, respectively. The presence of ester group was demonstrated by the carbon resonance at 165-172 ppm in the ^{13}C NMR spectrum, while the pyrimidine resonances were observed at: 58 (C-5), 150 (C-2) and 172 (C-4, C-6) ppm. Some of those compounds can serve useful monomers for synthesis of polyacrylates, crosslinking agents or plastificators of high thermal resistance due to the presence of pyrimidine rings.

Urethanes

The reactions of phenyl isocyanate with BHEDEB at 1:1 and 1:2 molar ratio in presence of triethylamine catalyst led to 2-[3-(2-hydroxyethyl)-2,4,6-trioxopyrimidyl]ethyl N-phenylcarbamate (**6, R = H**) or 2,2'-(5,5-diethyl-2,4,6-trioxopyrimidyl]diethyl bis(N-phenylcarbamate) (**7, R = H**) according to the following scheme:



The N-phenylcarbamates were not obtained when 1:1 or 1:2 BHPDEB and phenyl isocyanate were used for synthesis. Instead the triphenyl isocyanurate was obtained. When larger excess of isocyanate was applied, the 2,2'-(5,5-diethyl-2,4,6-trioxopyrimidyl)dipropyl bis(N-phenylcarbamates (**7, R = CH_3**) were obtained. The reason was probably the competing reaction of phenyl isocyanate cyclization into triphenyl isocyanurate and formation of diphenylurea according to the scheme:



This effective side reaction was presumably due to lower reactivity secondary hydroxyl groups present in BHPDEB with isocyanate groups in comparison with BHEDEB. The side products were unambiguously identified based on their IR, ^1H NMR, elemental analysis and melting points (see Analytical Data in Experimental). The separation of 7 (**R = CH_3**-) was laborious due to preceding selective precipitation of mentioned other products (see Procedure for the synthesis of urethane from BHPDEB in Experimental).

In the IR spectra of obtained phenylcarbamates the I and II amide bands were observed at 1668-1685 cm^{-1} and 1536-1546 cm^{-1} regions, respectively, as well as ester band at 1210 cm^{-1}. The band at ca. 1400 cm^{-1} showed clearly the presence of pyrimidine ring in products, while the band at 690-750 cm^{-1} indicated the presence of mono-substituted phenyl ring. In the spectra of products obtained from equimolar BHEDEB and phenyl isocyanate the deformation band of hydroxyl group was observed at ca. 1060 cm^{-1}. The ^{1}H NMR spectra of obtained urethanesters showed the methyl and methylene proton of pyrimidine resonances at 0.6-0.7 and 1.95 ppm, respectively, and methylene protons (in products obtained from BHEDEB) or methine protons (in products obtained from BHPDEB) linked to ester group (CH_2OCO or CHOCO) at 4.3 i 5.1-5.3 ppm, respectively. The ^{13}C-NMR indicated the ester carbon resonance at ca. 153 ppm, while pyrimidine resonances were identified at: 57 (C-2), 150 (C-5) and 172 (C-4, C-6) ppm. The phenyl carbon resonances were found at: 118. 3, 122.4 -123.4, 128.6 and 138.9.

Experimental

General procedure for the synthesis of esters

0.05 Moles of BHEDEB (13.6 g) or 1,3-bis(2-hydroxypropyl)-5,5-diethylbarbiturate (BHPDEB, 15.0 g) and 0.13 moles of an acid: acetic (7.8 g), methacrylic (11.9 g), acrylic (9.4 g) or stearic (28.4 g) were placed in three-necked 250 cm^3 flask equipped with Dean-Stark head and reflux condensed. Then 120 cm^3 of toluene solvent and 1.0-2.0 cm^3 concentrated sulfuric acid catalyst were added and the mixture was heated to reflux (110 - 115°C) and water was distilled off as azeotropic mixture with toluene. After the esterification reaction was completed the mixture was washed with 10 % aqueous potassium carbonate until evolution of gas ceased, then with water and finally the organic solvents were removed under vacuum to get resin product, which was further purified by high-vacuum distillation (t = 120-180°C, p = $0.7 \cdot 10^{-3}$ MPa = 5 mm Hg), or dried to constant mass in vacuum oven (t = 60°C, p = $2 \cdot 10^{-3}$ MPa = 15 mm Hg).

Additional remarks: (i) The reactions with acryclic acids were conducted in the presence of 2% (related to the mass of BHADEB) phenotiazine in order to prevent polymerization of unsaturated acids, (ii) The addition of potassium carbonate resulted in removal of sulfuric acid and unreacted acids used for esterification, (iii) Extraction with water led to removal of the formed salts and unreacted BHADEB.

Procedure for the synthesis of urethane from BHEDEB

0.02 Moles of BHEDEB (5.44 g) were added into 40 cm^3 dioxane freshly distilled from over sodium, in three-necked 100 cm^3 flask equipped with mechanical stirrer, thermometer and reflux condenser and the mixture was stirred until dissolving of reagents. Then 2.38 or 4.76 g (0.02 or 0.04 moles) of phenyl isocyanate and 0.05 cm^3 triethylamine (0.018 moles / mole of derivative of BHADEB) were added and the mixture was heated at 60 - 70°C. The heating was continued until the isocyanate number of mixture became zero.

Procedure for the synthesis of 2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)dipropyl bis(N-phenylcarbamate from BHPDEB

0.02 Moles of BHPDEB (6.0 g) were added into 40 cm^3 dioxane freshly distilled from over sodium, in three-necked 100 cm^3 flask equipped with mechanical stirrer, thermometer and reflux condenser nad the mixture was stirred until dissolving of

reagents. Then 14.4 g (0.12) of phenyl isocyanate and 0.05 cm^3 triethylamine (0.018 moles / mole of derivative of BHPDEB) were added and the mixture was heated at 60 - 70°C. The triphenyl isocyanate precipitated from reaction mixture upon standing for one day. The remaining filtrate was concentrated by half and further portion of triphenyl isocyanate was collected. The diphenylurea was precipitated from the filtrate upon addition of isopropyl alcohol. Then the isopropyl alcohol and dioxane were evaporated under reduced pressure. The resulting colorless oil crystallized upon standing one week in room temperature. The crude 2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)dipropyl bis(N-phenylcarbamate was further purified by crystallization from isopropyl alcohol.

Analyses

The progress of reaction between BHEDEB or BHPDEB and carboxylic acids was followed by determination of acidic number by by alkalimetric titration with 0.1 M KOH (aq) [14], while that of the reaction between BHADEB and phenyl isocyanate by determination of isocyanate by the ammonium method according to standard procedure [14]. The ^{1}H-NMR spectra of products were recoreded at 500 MHz Brucker UltraShield in DMSO-d_6 with hexamethylodisiloxane as internal standard. The IR spectra were taken with PARAGON 1000 FT Perkin Elmer spectrophotometer (Great Britain). Elemental analysis for C, H, N were done with EA 1108, Carlo-Erba (Italy) analyzer.

Analytical Data

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)diethyl diacetate (5, R = H, R' = CH_3), $C_{16}H_{24}N_2O_7$; Yield – 62%; n_D^{20} = 1.4800; elemental analysis - % Calcd.: C 53.93; H 6.74; N 7.87; % Found: C 53.54; H 6.88; N 7.84: IR (capillary film) [cm^{-1}], 2971-2881 (CH_3, CH_2), 1740 (C=O in ester), 1675 (C=O in pyrimidine ring), 1436 (CH_2), 1395 (C-N in ring), 1385 (CH_3), 1222 (CO-O), 1184 (C-N in ring), 1041 (C-O), 984, 805 (N-C=O): NMR (d_6-DMSO) [ppm]: 0.75 (6H, t, CH_2-C<u>H_3</u>), 1.85 (4H, m, C<u>H_2</u>-CH_3), 1.9 (6H, s OC-CH_3), 4.1 (4H, t, N-CH_2-), 4.3 (4H, t, CH_2-O); ^{13}C-NMR (d_6-DMSO) [ppm]: 8.8 (C5-CH_2-<u>C</u>H_3), 20.4 (<u>C</u>H_3-COO), 32.0 (C5-<u>C</u>H_2-CH_3), 43.3 (N-CH_2), 57.3 (C5), 60.7 (CH_2O), 150.3 (C2), 170.1 (COO), 171.1 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)diethyl dibenzoate (5, R = H, R' = Ph), $C_{26}H_{28}N_2O_7$; Yield – 45%; n_D^{20} = 1.5199; elemental analysis - % Calcd.: C 65.00; H 5.83; N 5.83; % Found: C 64.57; H 5.94; N 6.10; IR (capillary film) [cm^{-1}]: 3062-2808 (CH_3, CH_2, -CH= in Ar), 1716 (C=O in ester), 1676 (C=O in pyrimidine ring), 1435 (CH_2), 1384 (CH_3), 1267 (CO-O), 1175 (C-H in monosubst. Ar and C-N in pyrimidine ring), 1070 (C-O), 710 (C-H in monosubst. Ar); ^{1}H-NMR (d_6-acetone) [ppm]: 0.75 (6H, t, CH_2-C<u>H_3</u>), 1.95 (6H, q, C<u>H_2</u>-CH_3), 4.3 (4H, t, N-CH_2-), 4.5 (4H, t, CH_2-O), 7.5-8.0 (10H, m, Ar); ^{13}C-NMR (d_6-acetone) [ppm]: 9.5 (C5-CH_2-<u>C</u>H_3), 33.2 (C5-<u>C</u>H_2-CH_3), 41.5 (N-CH_2), 58.8 (C5), 62.8 (CH_2O), 127.8 (C3', C5'), 129.4 (C1'), 130.4 (C2', C6'), 133.7 (C4'), 151.8 (C2), 166.6 (COO), 172.3 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)diethyl distearate (5, R = H, R' = $(CH_2)_{16}CH_3$), $C_{48}H_{88}N_2O_7$; Yield – 35%; elemental analysis - % Calcd.: C 71.64; H 10.95; N 3.45; % Found: 71.55; H 10.80; N 3.51; IR (capillary film) [cm^{-1}]: 2954-2850 (CH_3, CH_2), 1747 (C=O in ester), 1689 (C=O in pyrimidine ring), 1472 (CH_2), 1397 (C-N in ring), 1268 (CO-O), 1088 (C-O), 988, 806 (N-C=O), 718 (CH_2); ^{1}H-NMR (d_6-acetone) [ppm]: 0.8 (6H, t, C_5-CH_2-C<u>H_3</u>), 0.9 (6H, t, $(CH_2)_{16}$-C<u>H_3</u>), 1.3 (28H, m, $(CH_2)_{14}$), 1.6 (4H, m, OCOCH_2C<u>H_2</u>), 1.95 (4H, q, C<u>H_2</u>-CH_3), 2.25 (4H, t, OCOC<u>H_2</u>CH_2), 4.2 (4H, t, N-CH_2-), 4.3 (4H, t, CH_2-O): ^{13}C-NMR (d_6-acetone)

[ppm]: 9.5 (C5-CH_2-$\underline{C}H_3$), 14.7 ((CH_2)$_{12}$-CH_2-CH_2-$\underline{C}H_3$), 23.3 ((CH_2)$_{12}$-CH_2-$\underline{C}H_2$-CH_3), 25.4 (-$\underline{C}H_2$-CH_2COO), 29.3-29.8 (($\underline{C}H_2$)$_{12}$-CH_2-CH_2-CH_3), 32.6 ((CH_2)$_{12}$-CH_2-$\underline{C}H_2$-CH_3), 33.4 ((CH_2)$_{12}$-$\underline{C}H_2$-CH_2-CH_3), 34.5 ((-CH_2-$\underline{C}H_2COO$), 41.5 (N-CH_2), 58.8 (C5), 61.7(CH_2O), 151.6 (C2), 172.3 (COO), 173.5 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)diethyl diacrylate (5, R = H, R' = CH=CH_2), $C_{18}H_{24}N_2O_7$; Yield – 60%; n_D^{20} = 1.4895; elemental analysis - % Calcd.: C 56.84; H 6.31; N 7.36; % Found: C 56.62; H 6.32; N 7.52; IR (capillary film) [cm^{-1}]: 3037 (CH_2=), 2971 (CH_2), 1725 (C=O in ester), 1675 (C=O in pyrimidine ring), 1635 (C=C), 1437 (CH_2), 1397 (C-N in ring), 1293, 1262 (CO-O), 1174 (C-N in ring), 1068 (C-O), 984, 808 (N-C=O); ^{1}H-NMR (d_6-acetone) [ppm]: 0.8 (6H, t, CH_2-$C\underline{H}_3$), 1.95 (4H, q, $C\underline{H}_2$-CH_3), 4.2 (4H, t, CH_2-O), 4.3 (4H, t, N-CH_2), 5,9 - 6,3 (CH=CH_2); ^{13}C-NMR (d_6-acetone) [ppm]: 8.9 (C5-CH_2-$\underline{C}H_3$), 32.1 (C5-$\underline{C}H_2$-CH_3), 43.3 (N-CH_2), 57.3 (C5), 61.1 (CH_2O), 127.9 (CH=), 129.4 (C1'), 131.7 (CH_2=), 150.2 (C2), 165.2 (COO), 171.0 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)diethyl dimethacrylate (5, R = H, R' = C(CH_3)=CH_2), $C_{20}H_{28}N_2O_7$; Yield – 54%; n_D^{20} = 1.4702; elemental analysis - % Calcd.: C 58.80; H 6.86; N 6,86; % Found: C 58.36; H 6.36; N 6.82; N 7.52; IR (capillary film) [cm^{-1}]: 2844-2966 (CH_3, -CH_2, CH_2=), 1720 (C=O in ester), 1686 (C=O in pyrimidine ring), 1634 (C=C), 1435 (CH_2), 1398 (C-N in ring), 1298 (CO-O), 1152 (C-O), 943 (=CH), 808 (N-C=O);^{1}H-NMR (d_6-acetone) [ppm]: 0.8 (6H, t, CH_2-$C\underline{H}_3$), 1.9 (6H, s, =C-CH_3), 1.95 (4H, m, $C\underline{H}_2$-CH_3), 4.2 (4H, t, N-CH_2-), 4.4 (4H, t, CH_2-O), 5.6, 6.1 (2H, 2d, =CH_2); ^{13}C-NMR (d_6-acetone) [ppm]: 9.5 (C5-CH_2-$\underline{C}H_3$), 18.4 (CH_3C=), 33.5 (C5-$\underline{C}H_2$-CH_3), 41.4 (N-CH_2), 58.8 (C5), 62.5 (CH_2O), 125.7 (CH_2=), 137.5 (C=), 133.7 (C4'), 151.7 (C2), 167.3 (COO), 172.2 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)dipropyl diacetate (5, R = R' = CH_3), $C_{18}H_{28}N_2O_7$; Yield – 50%; n_D^{20} =1.4805; elemental analysis - % Calcd. C 66.14; H 7.29; N 7,29; % Found: C 66.14; H 7.64; N 7.32; IR (capillary film) [cm^{-1}]: 3066 (CH_2=), 2966-2877 (CH_3, CH_2, CH), 1737 (C=O in ester), 1684 (C=O in pyrimidine ring), 1654 (C=C), 1543 (ring skeleton band), 1434 (CH_2), 1400 (C-N in ring), 1373 (CH_3), 1233 (CO-O), 1185 (C-N in ring) 1068, 1048 (C-O), 994, 831 (N-C=O); ^{1}H-NMR (d_6-DMSO) ([ppm]: 0.75 (6H, m, CH_2-$C\underline{H}_3$), 1.35 (6H, m, CH-$C\underline{H}_3$), 1.9 (10H, m, $C\underline{H}_2$-CH_3, OC-CH_3), 3.9, 4,2 (4H, m, N- CH_2); 5.1 (2H, m, CH-O); ^{13}C-NMR (d_6-DMSO) [ppm]: 8.4 (C5-CH_2-$\underline{C}H_3$), 17.4 (CH_3-$\underline{C}H$-O) 20.8 ($\underline{C}H_3$-COO), 31.7 (C5-$\underline{C}H_2$-CH_3), 45.0 (N-CH_2), 57.2 (C5), 67.9 (COO-$\underline{C}H$), 150.5 (C2), 169.9 (COO), 170.9 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)dipropyl dibenzoate (5, R = CH_3, R' = Ph), $C_{28}H_{32}N_2O_7$; Yield – 40%; n_D^{20} = 1.5278; elemental analysis - % Calcd. C 66.14; H 6.30; N 5.51; % Found: C 66.08; H 6.55; N 5.78; IR (capillary film) [cm^{-1}]: 2971-2880 (CH_3, CH_2, CH), 1716 (C=O in ester), 1682 (C=O in pyrimidine ring), 1435 (CH_2), 1400 (C-N i ring), 1267 (CO-O), 1177 (C-N in pyrimidine ring, C-H in monosubst. Ar), 1108 (C-O), 1070 (C-N in ring), 760, 709 (Ar-H); ^{1}H-NMR (d_6-acetone) [ppm]: 0.7 (6H, m, CH_2-$C\underline{H}_3$), 1.35 (6H, m, CH-$C\underline{H}_3$), 1.9 (4H, m, $C\underline{H}_2$-CH_3), 3.9, 4.2 (4H, m, N- CH_2), 5.3 (2H, m, CH-O), 7.5, 7.8 (10H, m, Ar-H); ^{13}C-NMR (d_6-acetone) [ppm]: 8.5 (C5-CH_2-$\underline{C}H_3$), 17.5 (CH_3-$\underline{C}H$-O) 31.7 (C5-$\underline{C}H_2$-CH_3), 45.4 (N-CH_2), 57.4 (C5), 69.1 (COO-$\underline{C}H$), 128.7 (C3', C5'), 129.9 (C1'), 130.3 (C2', C6'), 133.4 (C4'), 150.7 (C2), 165.4 (COO), 171.1 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)dipropyl diacrylate (5, R = CH_3, R' = CH=CH_2), $C_{20}H_{28}N_2O_7$; Yield – 49%; n_D^{20} =1.4989; elemental analysis - % Calcd. C 58.82; H 6.86; N 6.86; % Found: C 58.96; H 6.91; N 7.09; IR (capillary film) [cm^{-1}]: 2972-2881 (CH_3, CH_2, CH), 1722 (C=O in ester), 1681 (C=O in pyrimidine ring),

1638 (C=C), 1521 (ring skeleton band), 1435 (CH_2), 1400 (C-N in ring), 1383 (CH_3), 1291, 1266 (CO-O), 1191 (C-N in ring), 1067, 1050 (C-O), 985, 809 (N-C=O), 942 (CH_2=); ^{1}H-NMR (d_6-DMSO) [ppm]: 0.7 (6H, m, CH_2-C$\underline{H}_3$), 1.3 (6H, m, CH-C$\underline{H}_3$), 1.8 (4H, m, C$\underline{H}_2$-CH_3), 3.9, 4.2 (4H, m, N-CH_2), 5.1 (4H, m, CH-O and CH=), 5.9-6.2 (4H, m, CH_2=); ^{13}C-NMR (d_6-acetone) [ppm]: 8.7 (C5-CH_2-$\underline{C}H_3$), 17.5 (CH_3-$\underline{C}$H-O) 31.8 (C5-$\underline{C}H_2$-CH_3), 45.0 (N-CH_2), 57.2 (C5), 68.3 (COO-$\underline{C}$H), 128.2 (CH=), 131.6 (CH_2=), 150.4 (C2), 164.9 (COO), 170.9 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)dipropyl dimethacrylate (5, R = CH_3, R' = $C(CH_3)=CH_2$), $C_{22}H_{32}N_2O_7$; Yield – 48%; n_D^{20} =1.4740; elemental analysis - % Calcd. C 60.27; H 7.76; N 6.39; % Found: C 60.02; H 7.92; N 6.51; IR (capillary film) [cm^{-1}]: 3084 (CH_2=), 2966-2877 (CH_3, CH_2, CH), 1718 (C=O in ester), 1684 (C=O in pyrimidine ring), 1644 (C=C); 1529 (ring skeleton band), 1435 (CH_2), 1397 (C-N in ring), 1072, 1063 (C-O), 991 (N-C=O); 930 (CH_2=); ^{1}H-NMR (d_6-DMSO) [ppm]: 0.7 (6H, m, CH_2-C$\underline{H}_3$), 1.3 (6H, m, CH-C$\underline{H}_3$), 1.8 (10H, m, =C-CH_3, C$\underline{H}_2$- CH_3), 3.9, 4.2 (4H, m, N- CH_2), 5.1 (2H, m, CH-O), 5.1, 5.8 (4H, m, CH_2=); ^{13}C-NMR (d_6-DMSO) [ppm]: 8.9 (C5-CH_2-$\underline{C}H_3$), 17.5 ($\underline{C}H_3$-CH-O) 18.2 (CH_3C=), 32.0 (C5-$\underline{C}H_2$-CH_3), 43.3 (N-CH_2), 57.2 (C5), 68.3 (COO-$\underline{C}$H), 125.9 (CH_2=), 137.6 (C=), 150.0 (C2), 167.8 (COO), 170.6 (C4, C6).

2-[5,5-diethyl-3-(2-hydroxyethyl)-2,4,6-trioxopyrimidine]ethyl N-phenylcarbamate (6, R = H), $C_{19}H_{25}N_3O_6$, Yield – 54%; m.p =223-225^{0}C elemental analysis - % Calcd. C 58.31; H 6.39; N 10.74; % Found: C 58.02; H 6.35; N 10.52; IR (KBr) [cm^{-1}]: 3482-3346 (OH), 3199 (NH), 2969-2879 (CH_3;CH_2), 1732 (C=O in ester), 1668 (C=O in pyrimidine ring, and I amide bond), 1599 (NH), 1538 (II amide bond), 1440 (CH_2), 1395 (C-N in pyrimidine ring), 1390 (CH_3), 1311 (C-N), 1239 (III amide bond), 1210 (C-N in pyrimidine ring and CO-O), 1396 (CH_3), 1312 (C-N), 1064 (C-OH), 748, 708, 691 (CH in Ar); ^{1}H-NMR (d_6-DMSO), [ppm]; 0.7 (6H, t, CH_2-C$\underline{H}_3$); 1.9 (4H, q, C$\underline{H}_2$-CH_3); 3.5 (2H, t, CH_2-O), 3.9 (2H, t, N-CH_2), 4.3 (4H, t, CH_2-OCO), 4,8 (1H, t, OH), 7,0-7,5 (5H, m, Ar), 9,55 (1H, s, NH); ^{13}C-NMR(d_6-DMSO)[ppm]: 8.9 (C5-CH_2-$\underline{C}H_3$), 31.9 (C5-$\underline{C}H_2$-CH_3), 40.7 (N-$\underline{C}H_2CH_2OCO$), 43.2 (N-$\underline{C}H_2$-CH_2-OH) 57.5 (C5), 60.9 ($\underline{C}H_2OCO$), 66.3 (CH_2OH), 118. 3 (C2'), 122.4 (C4'), 128,6 (C3', C5'), 138.9 (C1'), 150.3 (C2), 153.3 (COO), 170.6 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)diethyl bis(N-phenylcarbamate) (7, R = H) $C_{26}H_{30}N_4O_7$; Yield – 92%; m.p =105-108^{0}C; elemental analysis - % Calcd. C 61.18; H 5.88; N 10.98; % Found: C 60.96; H 6.03; N 10.79; IR (capillary film) [cm^{-1}]:3356, 3306, 3173(NH), 2967-2877 (CH_3, CH_2), 1737 (C=O in ester), 1684 (C=O in pyrimidine ring and I amide bond), 1601 (NH), 1546 (II amide bond), 1486, 1455 (CH_2), 1402 (C-N in pyrimidine ring), 1390 (CH_3), 1311 (C-N), 1239 (III amide bond), 1210 (C-N in pyrimidine ring) and CO-O), 1090, 1063 (C-O), 749, 693 (CH w Ar); ^{1}H-NMR (d_6-DMSO), [ppm]; 0.6 (6H, t, CH_2-C$\underline{H}_3$), 1.9 (4H, q, C$\underline{H}_2$-CH_3), 4.1 (4H, t, N-CH_2), 4.3 (4H, t, CH_2-OCO), 7.0-7.5 (10H, m, Ar), 9.55 (2H, s, NH); ^{13}C-NMR (d_6-DMSO)[ppm]: 8.8 (C5-CH_2-$\underline{C}H_3$), 31.5 (C5-$\underline{C}H_2$-CH_3), 40.7 (N-$\underline{C}H_2CH_2OCO$), 57.2 (C5), 60.9 ($\underline{C}H_2OCO$), 118. 3 (C2'), 123.4 (C4'), 128.6 (C3', C5'), 138.9 (C1'), 150.3 (C2), 153.3 (COO), 170.9 (C4, C6).

2,2'-(5,5-diethyl-2,4,6-trioxopyrimidine)dipropyl bis(N-phenylcarbamate) (7, R = CH_3) $C_{28}H_{34}N_4O_7$; Yield 25%, m.p =23--231^{0}C; elemental analysis - % Calcd. C 60.27; H 7.76; N 6.39; % Found: C ; H ; N ; IR (KBr) [cm^{-1}]: 3245, 3193 (NH), 2993-2908 (CH_3, CH_2), 1727 (C=O in ester), 1707 (C=O in pyrimidine ring and I amide bond), 1603 (NH), 1560 (III amide bond), 1490, 1447 (CH_2), 1401 (C-N in pyrimidine ring), 1398 (CH_3), 1318 (C-N), 1238 (III amide bond), 1219 (C-N in

pyrimidine ring and CO-O), 1080, 1060 (C-O), 754, 745, 690 (CH w Ar); ^{1}H-NMR (d_6-DMSO) [ppm]: 0.7 (6H, t, CH_2-$C\underline{H_3}$), 1.3 (6H, d, CH-$C\underline{H_3}$), 1.9 (4H, q, $C\underline{H_2}$-CH_3), 3.9-4.2 (4H, d, N-CH_2), 4.9 (2H, m, CH-O), 7.0-7.4 (10H, m, Ar), 9.8 (1H, s, NH); ^{13}C-NMR (d_6-DMSO)[ppm]:

Triphenyl isocyanurate (8) $C_{21}H_{15}N_3O_3$; Yield 37%, m.p.$_{ref}$ = 285^{0}C [15], m.p. $_{found}$ = 285^{0}C; elemental analysis- % Calcd. C 70.58; H 4.20; N 11.76; % Found: C 70.56 ; H4.26 ; N 11.72; IR (KBr) [cm^{-1}]: 1710 (C=O in pyrimidine ring), 1510 (aromatic ring band) 1420 (C-N in pyrimidine ring), 1175 750, 700(C-H in Ph) ^{1}H-NMR (d_6-DMSO) [ppm]: 7.5 (15H, m, Ar).

Diphenylurea (9) $C_{13}H_{12}N_2O$; Yield 2%, m.p.$_{ref}$ = 239-241^{0}C [16], m.p.$_{found}$ = 238-241^{0}C; elemental analysis % Calcd. C 73.58; H 5.66; N 13.21; % Found: C 73.26 ; H 5.36 ; N 13.52; IR (KBr) [cm^{-1}]:3300 (NH), 3000 (C-H in Ar), 1650 (C=O),1590, 1550 (II amide bond), 1220 (III amide bond), 750, 680 (C-H in Ar); ^{1}H-NMR (d_6-DMSO) [ppm]: 6.9-7.5 (10H, m, Ar), 8,7 (2H, s, NH).

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