

Synthesis and Decomposition of Parabanic Acid Derivatives

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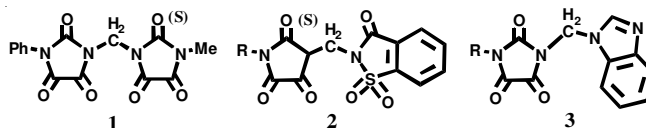
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As a part of a research program related to the synthesis of pharmacologically and agrochemically interesting parabanic acid derivatives, we synthesized imidazolidine-2,4,5-trionylimidazolidine (**6**), 2-thioxoimidazolidine-1-ylimidazolidine (**7**), benzo[d]-isothiazol-2-yl imidazolidine-2,4,5-thiones (**8a-c**), 2-thioxobenzo[d]-isothiazol-2-yl imidazolidine-4,5-dione (**9a-c**) and 1-benzimidazole-3-imidazolidine-2,4,5-triones (**10a-c**), 1-phenylurea (**11**), 1-ethyl-3-[(3-ethylthioureido)methyl]urea (**12**) and 1-ethyl-3-[(3-methylthioureido)methyl]urea (**13**) were obtained in basic solution through ultrasound.

Key Words: Parabanic acid, Benzimidazole, 1,2-Benzisothiazol-3-one-1,1-dioxide, Agrochemical fungicide, Urea derivatives.

INTRODUCTION

Parabanic acid(imidazolidine-2,4,5-trione, oxalylurea) resulting from the oxidation of biological fundamental compounds such as uric acid, guanine, uracil and alloxanic acid has attracted the attention of agricultural and medicinal chemists for many decades^{1,2}. Parabanic acid derivatives are useful pharmaceuticals for the treatment of diabetic complications such as diabetic neuropathy, diabetic cataracts and diabetic dermatopathy³⁻⁶. Further, parabanic acid derivatives are known for their herbicidal, plant growth regulating and fungicidal properties⁷. Polymers containing imidazolidine-2,4,5-trione (parabanic) rings are known as highly thermally stable polymers with improved chemical resistance in organic solvents⁸⁻¹³. Recently, many marine imidazol alkaloids have been isolated from sponges and their antitumor and antibacterial activities have also been identified^{14,15}. In our previous works to obtain new agrochemicals, we reported the synthesis of new saccharin derivatives containing the 2,4,5-imidazolidinetrione group¹⁶. As a part of research program related to the synthetic study of pharmacologically and agrochemically important imidazolidines¹⁶, we chose to identify imidazolidine-2,4,5-trione and imidazolidine-2,4,5-trione,2-thioxo-imidazolidine-4,5-dione, benzimidazole, orsaccharin rings as active components for the desired property, the synthesis of (imidazolidine-2,4,5-trionyl) imidazolidine-2,4,5-triones (**1**), 2,4,5-imidazolidinetrionyl-1,2-benzisothiazol-3-one-1,1-di-oxide **2**, 4,5-dioxo-2-thioxo-imidazolidin-1-ylmethyl-imidazolidine-2,4,5-triones (**2**) and 1-benzimidazole-1-yl-3-imidazolidine-2,4,5-trione (**3**) are already reported.



EXPERIMENTAL

Melting points were determined on a Büchi 510 capillary melting point apparatus and uncorrected. Infrared spectra were recorded on a Perkin-Elmer 683 spectro-photometer. NMR spectra were recorded on a Varian XL-300 or Bruker AC 200 FT-NMR spectrometer in CDCl₃ containing Me₄Si as an internal reference. Mass spectra were obtained by using JEOL JMS DX 303 or HP 5892 Mass Spectrometer.

General procedure for the synthesis of **6,7:** To a solution of 1-chloromethyl-imidazolidine-2,4,5-triones (**4**) (or 1-chloromethyl-2-thioxo-imidazolidine-4,5-diones (**5**), 1.13×10^{-2} mol) in dry THF (15 mL) under nitrogen at room temperature was added solution of 2,4,5-imidazolidinetriones (or 2-thioxo-imidazolidine-4,5-diones, 1.13×10^{-2} mol) and triethylamine (0.4 mL) in dry THF (15 mL). The reaction mixture was stirred at room temperature for 0.5 h. After 0.5 h, the reaction mixture was refluxed at 60-65 °C for 7 h. The reaction mixture was cooled again to room temperature and THF (75 mL) was added. The combined organic layers were dried over anhydrous Na₂SO₄, filtered and concentrated *in vacuo*. The residue was purified by flash column chromatography on silica gel eluted with only CH₂Cl₂, to provide products **6,7**.

1-[(3-Phenylimidazolidine-2,4,5-trionyl)methyl]-3-methyl-imidazolidine-2,4,5-trione (6**):** m.p. 247-249 °C;

^1H NMR (200 MHz, DMSO) 3.08 (s, 3H), 5.43 (s, 2H), 7.52 (m, 5H); ^{13}C NMR (50 MHz, DMSO) 157.2, 156.1, 155.9, 155.7, 152.8, 151.7, 130.2, 129.3, 128.9, 126.5, 40.6, 24.5; GC/MS m/z 330 ($[\text{M}]^+$); Anal. calcd. for $\text{C}_{14}\text{H}_{10}\text{N}_4\text{O}_6$: C, 50.92; H, 3.05; N, 16.96. Found : C, 50.95; H, 3.04; N, 16.94.

1-[(4,5-Dioxo-3-phenyl-2-thioxoimidazolidin-1-yl)methyl]-3-ethyl-imidazolidine-2,4,5-trione (7): m.p. 217–218 °C; ^1H NMR (200 MHz, DMSO) 1.11 (t, $J = 7.1$ Hz, 6H), 3.51 (d, $J = 6.9$ Hz, 2H), 5.53 (s, 2H), 3.80 (d, $J = 7.0$ Hz, 2H); ^{13}C NMR (50 MHz, DMSO) 180.1, 156.7, 156.0, 154.9, 154.7, 152.4, 43.6, 36.6, 33.8, 12.9, 12.5; GC/MS m/z 360 ($[\text{M}]^+$); Anal. calcd. for $\text{C}_{15}\text{H}_{12}\text{N}_4\text{O}_5\text{S}$: C, 50.00; H, 3.36; N, 15.55. Found : C, 50.03; H, 3.35; N, 15.59.

The typical experimental procedure for 1-phenyl-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-2,4,5-trione (**6**) is as follows: To a solution of 1-chloromethyl-3-phenylimidazolidine-2,4,5-trione (**4c**) (2.38 g, 10 mmol) in dry THF (15 mL) under nitrogen at room temperature was added solution of saccharin (2.01 g, 11 mmol) and triethylamine (1.2 mL) in dry THF (15 mL). The reaction mixture was stirred at room temperature for 0.5 h. After 0.5 h, the reaction mixture was refluxed at 55–60 °C for 5 h. The reaction mixture was cooled again to room temperature and THF (50 mL) was added. The combined organic layers were dried over anhydrous Na_2SO_4 , filtered and concentrated *in vacuo*. The residue was purified by flash column chromatography on silica gel eluted with only CH_2Cl_2 , to provide the 1-phenyl-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-2,4,5-trione **6** as a white crystalline solid (1.93 g, 50 %). m.p. 191–192 °C; ^1H NMR (200 MHz, CDCl_3) 5.84 (s, 2H, CH_2), 7.4–8.1 (m, 9H, phenyl); Mass m/z (rel. intensity, %) 385 ($[\text{M}]^+$, 40), 196 (100), 91, 77.

The typical experimental procedure for 1-ethyl-2-thioxo-3-(1,1,3-tri-oxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-4,5-dione (**7**) is as follows: To a solution of 1-ethyl-2-thioxo-imidazolidine-4,5-dione (**5b**) (1 g, 4.8 mmol) in the dry THF (10 mL) under nitrogen at room temperature was added solution of saccharin (1.5 g, 8.2 mmol) and triethylamine (1.1 mL) in the dry THF (10 mL). The reaction mixture was stirred at room temperature for 0.5 h. After 0.5 h, the mixture was refluxed 55–60 °C for 5 h. The reaction mixture was cooled again to room temperature and THF (50 mL) was added. The combined organic layers were dried over anhydrous Na_2SO_4 , filtered and concentrated *in vacuo*. The residue was purified by flash column chromatography on silica gel eluted with only CH_2Cl_2 , to provide the 1-ethyl-2-thioxo-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-4,5-dione (**7**) as a yellow crystalline solid (1.68 g, 49 %). m.p. 157–158 °C; ^1H NMR (200 MHz, acetone- d_6) 1.17 (t, 3H, $-\text{CH}_3$), 3.99–4.06 (q, 2H, $-\text{CH}_2$), 6.04 (s, 2H, N- CH_2 -N), 8.05–8.20 (m, 4H, phenyl); IR (KBr, ν_{max} , cm^{-1}): 1780, 1740, 1720, 1390, 1330, 1280, 1240, 1175; Mass m/z (rel. intensity, %) 352 ($[\text{M}]^+$).

1-Chloromethyl-3-methyl-imidazolidine-2,4,5-trione (4a): Yield 50 %; m.p. 149–150 °C; IR (KBr, ν_{max} , cm^{-1}): 1731, 1459, 1407, 1306, 1129; Mass m/z (rel. intensity, %) 179 ($[\text{M}+2]^+$, 5), 176 ($[\text{M}]^+$, 20), 141 (100), 113 (30), 94 (5), 70 (35), 56 (85); ^1H NMR (200 MHz, CDCl_3) 3.23 (s, 3H, CH_3), 5.40 (s, 2H, CH_2Cl).

1-Chloromethyl-3-ethyl-imidazolidine-2,4,5-trione (4b): Yield 64 %; m.p. 83–84 °C; IR (KBr, ν_{max} , cm^{-1}) 2982, 2865, 1735, 1409, 1298, 1208, 1128; Mass m/z (rel. intensity, %) 192 ($[\text{M}+1]^+$, 5), 190 ($[\text{M}-1]^+$, 10), 175 (2), 162 (1), 154 (30), 127 (15), 99 (10), 70 (45), 56 (100); ^1H NMR (200 MHz, CDCl_3) 1.27–1.34 (t, 3H, CH_3), 3.70–3.81 (q, 2H, CH_2Cl), 5.39 (s, 2H, CH_2Cl).

1-Chloromethyl-3-phenyl-imidazolidine-2,4,5-trione (4c): Yield 70 %; m.p. 130–131 °C; IR (KBr, ν_{max} , cm^{-1}) 1780, 1730, 1500, 1440, 1295, 1190; Mass m/z (rel. intensity, %) 238 ($[\text{M}]^+$, 27), 119 (100), 91 (23); ^1H NMR (200 MHz, CDCl_3) 5.6 (s, 2H, CH_2Cl), 7.4–7.5 (m, 5H, phenyl).

1-Methyl-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-2,4,5-trione (8a): Yield 40 %; m.p. 192–193 °C; IR (KBr, ν_{max} , cm^{-1}): 1749, 1453, 1340, 1291, 1245, 1179; Mass m/z (rel. intensity, %) 323 ($[\text{M}]^+$, 1), 259 (15), 223 (4), 196 (100), 174 (20), 169 (17), 132 (20), 121 (5), 104 (22), 76 (15), 70 (9); ^1H NMR (200 MHz, Acetone- d_6) 3.14 (s, 3H, CH_3), 5.70 (s, 2H, CH_2), 8.05–8.21 (m, 4H, phenyl).

1-Ethyl-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-2,4,5-trione (8b): Yield 54 %; m.p. 182–183 °C; IR (KBr, ν_{max} , cm^{-1}): 2981, 2885, 1746, 1425, 1340, 1293, 1251, 1180, 1115; Mass m/z (rel. intensity, %) 337 ($[\text{M}]^+$, 2), 273 (8), 223 (4), 196 (100), 174 (15), 169 (14), 132 (20); ^1H NMR (200 MHz, Acetone- d_6) 1.18–1.28 (t, 3H, CH_3), 3.64–3.75 (q, 2H, CH_2), 5.70 (s, 2H, N- CH_2 -N), 8.02–8.22 (m, 4H, phenyl).

1-Phenyl-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-2,4,5-trione (8c): Yield 50 %; m.p. 191–192 °C; IR (KBr, ν_{max} , cm^{-1}): 1785, 1750, 1410, 1330, 1290, 1250; Mass m/z (rel. intensity, %) 385 ($[\text{M}]^+$, 40), 196 (100), 169 (20), 119 (80), 91 (45), 77 (33); ^1H NMR (200 MHz, CDCl_3) 5.84 (s, 2H, CH_2), 7.4–8.1 (m, 9H, phenyl).

1-Chloromethyl-3-methyl-2-thioxo-imidazolidine-4,5-dione (5a): Yield 45 %; m.p. 97–98 °C; IR (KBr, ν_{max} , cm^{-1}): 1784, 1772, 1405, 1378, 1354; Mass m/z (rel. intensity, %) 192 ($[\text{M}]^+$, 1); ^1H NMR (200 MHz, Acetone- d_6) 3.38 (s, 3H, CH_3), 5.76 (s, 2H, CH_2).

1-Chloromethyl-3-ethyl-2-thioxo-imidazolidine-4,5-dione (5b): Yield 45 %; m.p. 108–109 °C; IR (KBr, ν_{max} , cm^{-1}): 1780, 1740, 1402, 1370, 1230, 1135; Mass m/z (rel. intensity, %) 206 ($[\text{M}]^+$); ^1H NMR (200 MHz, Acetone- d_6) 1.21–1.28 (t, 3H, CH_3), 3.95–4.06 (q, 2H, CH_2), 5.75 (s, 2H, CH_2Cl).

1-Chloromethyl-3-phenyl-2-thioxo-imidazolidine-4,5-dione (5c): Yield 40 %; m.p. 149–150 °C; IR (KBr, ν_{max} , cm^{-1}): 1780, 1650, 1520, 1500, 1400, 1230; Mass m/z (rel. intensity, %) 254 ($[\text{M}]^+$); ^1H NMR (200 MHz, Acetone- d_6) 5.87 (s, 2H, CH_2Cl), 7.43–7.60 (m, 5H, phenyl).

1-Methyl-2-thioxo-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-4,5-dione (9a): Yield 40 %; m.p. 173–174 °C; IR (KBr, ν_{max} , cm^{-1}): 1780, 1400, 1340, 1290, 1250, 1170, 1100; Mass m/z (rel. intensity, %) 339 ($[\text{M}]^+$); ^1H NMR (200 MHz, Acetone- d_6) 3.39 (s, 3H, CH_3), 6.04 (s, 2H, N- CH_2 -N), 8.10–8.17 (m, 4H, phenyl).

1-Ethyl-2-thioxo-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-4,5-dione (9b): Yield 49 %; m.p. 157–158 °C; IR (KBr, ν_{max} , cm^{-1}): 1780, 1740, 1720, 1390, 1330, 1280, 1240, 1175; Mass m/z (rel. intensity, %) 353 ($[\text{M}]^+$); ^1H NMR (200 MHz, Acetone- d_6)

1.17 (s, 3H, CH₃), 3.99-4.06 (s, 2H, CH₂), 6.04 (s, 2H, N-CH₂-N), 8.05-8.20 (m, 4H, phenyl).

1-Phenyl-2-thioxo-3-(1,1,3-trioxo-1,3-dihydro-16-benzo[d]isothiazol-2-ylmethyl)-imidazolidine-4,5-dione (9c): Yield 30 %; m.p. 170-171 °C; IR (KBr, $\nu_{\max}$, cm⁻¹): 1785, 1720, 1400, 1370, 1360, 1300, 1250, 1180; Mass m/z (rel. intensity, %) 402 ([M]⁺); ¹H NMR (200 MHz, acetone-*d*₆) 6.58 (s, 2H, CH₂), 7.84-8.63 (m, 9H, phenyl).

1-Benzoimidazole-1-yl-methyl-3-imidazolidine-2,4,5-trione (10a): R_f 0.7 (TLC eluent; *n*-Hexane : EtOAc = 1 : 25, v/v); yield 75 %; m.p. 188-189 °C; IR (KBr, $\nu_{\max}$, cm⁻¹): 3846, 1778, 1452, 1344, 1244, 756; Mass m/z (rel. intensity, %) 258 (100), 173 (7), 145 (9), 131 (100), 118 (15), 104 (12), 90 (15), 77 (20), 56 (64), 51 (12); ¹H NMR (200 MHz, CDCl₃) 3.18 (s, 3H, CH₃), 6.18 (s, 2H, N-CH₂-N), 7.25-7.78 (m, 4H, phenyl), 8.2 (s, 1H, benzimidazole).

1-Benzoimidazole-1-yl-ethyl-3-imidazolidine-2,4,5-trione (10b): R_f 0.7 (TLC eluent; *n*-Hexane: EtOAc = 1 : 25, v/v); yield 50 %; m.p. 179-180 °C; IR (KBr, $\nu_{\max}$, cm⁻¹): 3541, 2984, 1736, 1425, 1242, 796; Mass m/z (rel. intensity, %) 272 (93), 245 (2), 173 (5), 145 (7), 131 (100), 118 (11), 104 (8), 90 (10), 77 (14), 56 (57), 51 (8); ¹H NMR (200 MHz, CDCl₃) 1.25 (t, 3H, CH₃), 3.65 (q, 2H, CH₂), 5.97 (s, 2H, N-CH₂-N), 7.25-7.82 (m, 4H, phenyl), 8.19 (s, 1H, benzimidazole).

1-Benzoimidazole-1-yl-phenyl-3-imidazolidine-2,4,5-trione (10c): R_f 0.7 (TLC eluent; *n*-Hexane : EtOAc = 1 : 25, v/v); yield 45 %; m.p. 187-188 °C; IR (KBr, $\nu_{\max}$, cm⁻¹) 3884, 2924, 1767, 1741, 1242; Mass m/z (rel. intensity, %) 320 (61), 173 (5), 145 (4), 131 (100), 119 (7), 91 (8), 77 (12), 64 (6), 56 (26); ¹H NMR (200 MHz, CDCl₃) 6.15 (s, 2H, N-CH₂-N), 7.15-7.85 (m, 9H, phenyl), 8.25 (s, 1H, benzimidazole).

1-Phenylurea (11): Isolated yield 41%; R_f 0.35 (TLC eluent; EtOAc : *n*-Hexane = 1 : 5, v/v); Mass m/z (rel. intensity, %) 135; ¹H NMR (200 MHz, CDCl₃) 6.20 (s, 1H), 7.19 (m, 1H), 7.43 (m, 2H), 7.61 (dd, 2H), 8.9 (s, 1H); ¹³C NMR (50 MHz, CDCl₃) 121.6, 128.0, 128.9, 139.4; Anal. calcd. for C₇H₈N₂O : C, 61.75; H, 5.92; N, 3.47; Found : C, 61.73; H, 5.91; N, 3.44.

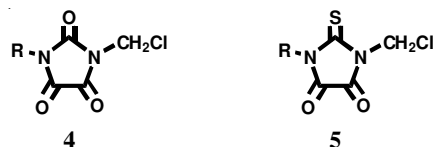
1-Ethyl-3-[(3-ethylthioureido)methyl]urea (12): Isolated yield 26 %; R_f 0.35 (TLC eluent; EtOAc : *n*-Hexane = 1 : 8, v/v); Mass m/z (rel. intensity, %) 204 (29), 167 (29), 149 (100), 104 (43), 76 (38), 70 (55), 55 (100); ¹H NMR (200 MHz, CDCl₃) 6.20 (s, 1H), 7.19 (m, 1H), 7.43 (m, 2H), 7.61 (dd, 2H), 8.9 (s, 1H); ¹³C NMR (50 MHz, CDCl₃) 14.5, 15.3, 34.2, 40.2, 62.3, 157.9, 182.2; Anal. calcd. for C₇H₁₆N₄O-S : C, 41.15; H, 7.89; N, 27.40; O, 7.83; S, 15.7; Found: C, 41.13; H, 7.88; N, 27.40; O, 7.79; S, 15.64.

1-Ethyl-3-[(3-methylthioureido)methyl]urea (13): Isolated yield 23 %; R_f 0.35 (TLC eluent; EtOAc: *n*-Hexane = 1 : 8, v/v); Mass m/z (rel. intensity, %) 190 (5), 119 (100), 91 (95), 77 (5), 64 (81), 51 (31); ¹H NMR (200 MHz, CDCl₃) 6.20 (s, 1H), 7.19 (m, 1H), 7.43 (m, 2H), 7.61 (dd, 2H), 8.9 (s, 1H); ¹³C NMR (50 MHz, CDCl₃) 14.5, 31.0, 34.3, 62.3, 157.7, 182.2; Anal. calcd. for C₆H₁₄N₄O-S : C, 37.88; H, 7.42; N, 29.45; O, 8.41; S, 16.85; Found: C, 37.84; H, 7.40; N, 29.41; O, 8.40; S, 16.82.

RESULTS AND DISCUSSION

The basic catalyzed condensation between imidazolidine-2,4,5-triones and paraformaldehyde in aqueous solution

yielded a mixture of the expected *N*-hydroxy methyl derivatives and imidazolidine-2,4,5-triones. However, the instability of *N*-hydroxy methyl derivatives made its isolation very difficult. The use of column chromatography as a method of purification was not successful, whatever the eluent or support (silica gel, alumina) was used, because the R_f value was the same for the two compounds. For this reaction, the next chlorination step, using a large excess of thionyl chloride, was realized starting directly from a mixture of *N*-hydroxy methyl derivatives and imidazolidine-2,4,5-triones. The chlorinated precursors **4,5** were easily isolated by column chromatography and the product had a much higher R_f value than the starting material.



When the chlorinated products **4** and **5** were allowed to react with *N*-methylimidazolidine-2,4,5-trione and *N*-phenylimidazolidine-2,4,5-trione, respectively, 1-[(3-phenylimidazolidine-2,4,5-trionyl)methyl]-3-methyl-imidazolidine-2,4,5-trione **6** and 1-[(4,5-dioxo-3-phenyl-2-thioxoimidazolidin-1-yl)methyl]-3-ethylimidazolidine-2,4,5-trione **7** were obtained in good yields, as shown in Table-1. When chlorinated products **4** and **5** were allowed to react with 1,2-benzothiazole-3-one-1,1-dioxide by same methods, saccharin derivatives **8,9** containing imidazolidine-2,4,5-trionyl, 2-thioxo-imidazolidine-4,5-dionyl groups were obtained in good yields as shown in the Table-1. We also tried to obtain various parabanic acid derivatives from the reactions of the chlorinated reactants **4,5** with benzimidazole. 1-Benzoimidazole-1-yl-methyl-3-imidazolidine-2,4,5-trione **10a** (yield, 75 %), 1-benzoimi-

TABLE-1
YIELDS OF PRODUCTS **6-10**

Entry	Product	Yield ^a (%)	m.p. (°C)	
1	 6	76	247-249	
2	 7	69	217-218	
3	 8a-c	R; a = Me	52	192-193
		b = Et	54	182-183
		c = Ph	50	191-192
4	 9a-c	R; a = Me	40	173-175
		b = Et	49	157-159
		c = Ph	30	191-192
5	 10a-c	R; a = Me	75	188-189
		b = Et	50	179-180
		c = Ph	45	187-188

^aIsolated yields

^aIsolated yields

dazole-1-yl-ethyl-3-imidazolidine-2,4,5-trione (**10b**) (yield, 50 %) and 1-benzimidazole-1-yl-phenyl-3-imidazolidine-2,4,5-trione (**10c**) (yield, 45 %) were obtained in good yields, as shown in Table-1.

Biological tests for testing the phytocidal, herbicidal and insecticidal properties of the new synthesized compounds (**6-10**) are currently in progress. As a part of a research program related to the synthesis study of bioactive organo chemicals, we also report the synthesis of Di-urea derivatives in the reaction of 1-phenylimidazolidine-2,4,5-trione, 1-ethyl-3-[(3-ethyl-4,5-dioxo-2-thioxoimidazolidin-1-yl)methyl]-imidazolidine-2,4,5-trione and 1-ethyl-3-[(3-methyl-4,5-dioxo-2-thioxoimidazolidin-1-yl)methyl]-imidazolidine-2,4,5-trione with hydrolysis in basic solution through ultrasound.

1-Phenylurea (**11**), 1-ethyl-3-[(3-ethylthioureido)methyl]-urea (**12**) and 1-ethyl-3-[(3-methylthioureido)methyl]urea (**13**) were obtained in good yields as shown in Table-2.

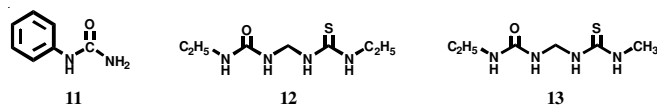


TABLE-2
YIELDS OF OBTAINED PRODUCTS **11**,
12, **13** THROUGH ULTRASOUND

Entry	Product	Reaction time (h)	Yield* (°C)
1		20	41
2		8	26
3		8	23

*Isolated yield

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