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# FACILE OXIDATION OF 2-OXO-1,4,6-TRISUBSTITUTED 1,2,3,6-TETRAHYDROPYRIMIDINES WITH CHLORANIL

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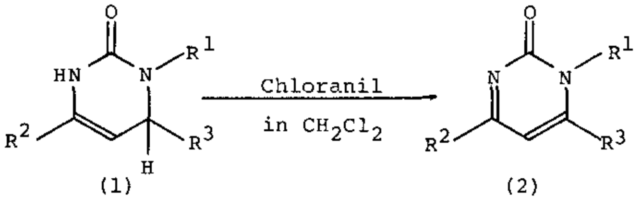
Abstract 2-Oxo-1,4,6-trisubstituted 1,2,3,6-tetrahydropyrimidines (**1a**-**1h**) were easily oxidized with 2,3,5,6-tetrachloro-1,4-benzoquinone(chloranil) to afford the corresponding 1,4,6-trisubstituted 2(1H)-pyrimidinones (**2**) in good yields.

2(1H)-Pyrimidinones have been supplied by the reaction of ureas with acetylacetone$^{1)}$, the condensation of acetylenic ketones with ureas$^{2)}$, the reaction of diimines with ethyl chloroformate.$^{3)}$ However, the only two attempts to prepare 2(1H)-pyrimidinones by oxidation of 2-oxo-tetrahydropyrimidines with potassium permanganate and manganese dioxide were carried out by Hardtmann$^{4)}$ and Coppola$^{5)}$, respectively, to the best of our knowledge.

In this paper, we wish to report on the reaction of 2-oxo-1,4,6-trisubstituted 1,2,3,6-tetrahydropyrimidines with various oxidizing agents. 2-Oxo-1,4,6-trisubstituted 1,2,3,6-tetrahydropyrimidines (**1**) were prepared from the reduction of 1,4,6-trisubstituted 2(1H)-pyrimidinones (**2**) with sodium borohydride in the presence of trimethyl borate. 2-Oxo-1-phenyl-4,6-dimethyl-1,2,3,6-tetrahydropyrimidine (**1a**) was treated with manganese dioxide or potassium ferricyanide to recover the starting material. Further, we tried oxidation of **1a** with potassium permanganate or potassium permanganate in the presence of phase transfer catalysis, $PhCH_2\overset{+}{N}Et_3Cl^-$, to give intractable mixture in both cases. On the contrary, **1a** was smoothly oxidized with chloranil to afford the corresponding 1-phenyl-4,6-dimethyl-2(1H)-pyrimidinone (**2a**) in 96% yield. The oxidation was carried out as follows. Chloranil (1.2 mmol) was added to the solution of 2-oxo-tetrahydropyrimidine (**1a**, 1 mmol) in dichloromethane (20 ml). The mixture was stirred for an hour at room temperature and the yield was determined by lpc. The oxidation of other 2-oxo-tetrahydropyrimidines (**1b**-**1h**) was

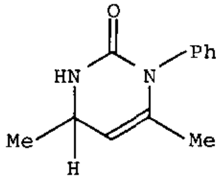
Table 1



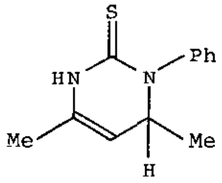
(1) (2)

| | $R^1$ | $R^2$ | $R^3$ | Yield(%)* |
|---|---|---|---|---|
| a | $C_6H_5$- | $CH_3$- | $CH_3$- | 96 |
| b | p-$CH_3C_6H_4$- | $CH_3$- | $CH_3$- | 94 |
| c | m-$CH_3OC_6H_4$- | $CH_3$- | $CH_3$- | 98 |
| d | m-$ClC_6H_4$- | $CH_3$- | $CH_3$- | 92 |
| e | m-$CH_3C_6H_4$- | $CH_3$- | $CH_3$- | 96 |
| f | $C_6H_5$- | $C_6H_5$- | $CH_3$- | 99 |
| g | $C_6H_5$- | $CH_3$- | $C_6H_5$- | 88 |
| h | $CH_3$- | $CH_3$- | $C_6H_5$- | 88 |

*Determined by lpc.



(3)



(4)

performed in the same manner as described above and the yields were summarized in Table 1. 2-Oxo-1-phenyl-4,6-dimethyl-1,2,3,4-tetrahydropyrimidine (3) was also oxidized with chloranil to yield 2(1H)-pyrimidinone (2a), but the oxidation product couldn't be obtained in the case of 2-thioxo-1-phenyl-4,6-dimethyl-1,2,3,6-tetrahydropyrimidine (4).

From these results, we found that 2-oxo-1,4,6-trisubstituted 1,2,3,6-tetrahydropyrimidines were easily oxidized with chloranil in high yields.

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