Reaction of Pyrimidin-2(1*H*)-ones and -thiones with Organometallic Compounds. Regioselective Preparation of Dihydropyrimidin-2(1*H*)-ones and -thiones

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4,6-Dimethyl-1-phenylpyrimidin-2(1*H*)-one (1) reacted with methylmagnesium iodide to afford 4,6,6-trimethyl-1-phenyl-3,6-dihydropyrimidin-2(1*H*)-one (8a) selectively; compound (1) reacted with methyl-lithium to give mainly 4,4,6-trimethyl-1-phenyl-3,4-dihydropyrimidin-2(1*H*)-one (8b). The reactions of various pyrimidin-2(1*H*)-ones and -thiones with organometallic compounds, and the influence of bulkiness of alkyl groups upon the product ratio, are also discussed.

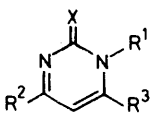
VARIOUS dihydropyrimidin-2(1*H*)-ones and -thiones have been prepared from the reaction of 1,1-dimethyl-3-oxobutyl isocyanate [1] and 2-methyl-2-thiocyanopentan-4-one [2] with anilines, αβ-unsaturated enones [3-5] and 3-ethoxy-2-methoxymethylenepropionitrile with ureas,[6,7] and the condensation of mesityl oxide, ammonium thiocyanate, and amines.[8] On the other hand, few papers concerning the preparation of dihydropyrimidin-2(1*H*)-ones and thiones by the reaction of pyrimidin-2(1*H*)-ones and -thiones with nucleophiles have been reported.[9]

The 2-, 4-, and 6-position atoms in the pyrimidine ring are electron deficient by virtue of the electron-withdrawing effect of the ring-nitrogen atoms.[10] Nucleophilic attack, therefore, could be expected to take place on these carbon atoms. While Hauser and co-workers reported that organolithium reagents reacted with benzalacetophenone to yield predominantly 1,2-addition product, the Grignard reagents gave exclusively the 1,4-addition product.[11]

In this paper, we discuss the regioselective preparation of dihydropyrimidin-2(1*H*)-ones and -thiones by the reaction of pyrimidin-2(1*H*)-ones and thiones with organometallic compounds such as Grignard and organolithium reagents.

RESULTS AND DISCUSSION

The reaction of 4,6-dimethyl-1-phenylpyrimidin-2(1*H*)-one (1) with methylmagnesium iodide (MeMgI) yielded two products, m.p. 191—192 °C (compound A) and m.p. 163 °C (compound B). Both compounds had the same formula, $C_{13}H_{16}N_2O$, from elemental analysis. Compound A shows the following spectral data; i.r., $\nu_{max.}$ 3 200 (N–H) and 1 660 cm^{-1} (C=O), ^1H n.m.r., δ 1.18 (6 H, s) and 1.70 (3 H, d, J = 0.6 Hz), attributed to the allylic coupling of methyl protons with the olefinic proton at 5-position of the pyrimidine ring. Compound B; i.r. $\nu_{max.}$ 3 220 (N–H) and 1 660 cm^{-1} (C=O); and ^1H n.m.r., δ 1.30 (6 H, s) and 1.53 (3 H, d, J = 0.6 Hz). From these spectral data, compounds A and B were assumed to be structurally isomeric with each other, and compound B was found to be 4,4,6-trimethyl-1-phenyl-3,4-dihydropyrimidin-2(1*H*)-one (8b) by comparison with an authentic sample (m.p. 162—164 °C) obtained from the reaction of 1,1-dimethyl-3-oxobutyl isocyanate with aniline.[1] It appears that compound (8a) is formed by the attack of the methyl Grignard at the 4-position of the pyrimidine ring. Therefore, compound A was assigned the structure 4,6,6-trimethyl-1-phenyl-3,6-dihydropyrimidin-2(1*H*)-one (8a) which was formed by the attack of the methyl Grignard at the 6-position. The total yield of (8a) and (8b) was 29%, in the ratio of 95 : 5. Pyrimidin-2(1*H*)-one (1) also reacted with methyl-lithium (MeLi) to give two products (8a) and (8b) in 65% yield, but the ratio was 15 : 85 (see Table).



(1) X = O, R^1 = Ph, R^2 = R^3 = Me
(2) X = S, R^1 = Ph, R^2 = R^3 = Me
(3) X = O, R^1 = *p*-MeC_6H_4, R^2 = R^3 = Me
(4) X = O, R^1 = Me, R^2 = R^3 = H
(5) X = O, R^1 = R^2 = Ph, R^3 = Me
(6) X = S, R^1 = Ph, R^2 = Me, R^3 = Ph
(7) X = O, R^1 = Ph, R^2 = R^3 = H

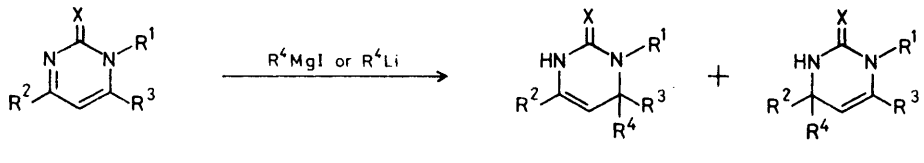
When 4,6-dimethyl-1-phenylpyrimidin-2(1*H*)-thione (2) was treated with MeMgI and MeLi to afford a mixture of two products, (12a) and (12b), in 62 and 43% yield, respectively. The structures of (12a) and (12b) were determined from the fact that the chemical shifts of the geminal dimethyl and allylic methyl protons were very similar to those of compounds (8a) and (8b), respectively. Further, (12b) was found to be identical with an authentic sample of 4,4,6-trimethyl-1-phenyl-3,4-dihydropyrimidine-2(1*H*)-thione (12b) obtained from the reaction of 2-methyl-2-thiocyanatopentan-4-one with aniline.[8] Similarly, the reaction of pyrimidin-2(1*H*)-one (1) and -thione (2) with various alkyl Grignards and alkyllithium reagents was examined and the results are summarized in the Table.

These results suggest that the bulkiness of the alkyl group of the Grignard reagent has a large influence on the ratio of the 3,6-dihydropyrimidin-2(1*H*)-ones (8a), (9a), (11a) to the 3,4-dihydropyrimidin-2(1*H*)-ones (8b)—(11b), but not on the ratio of the 2-thioxo-derivatives (12a)—(15a) to the corresponding derivatives (12b)—(15b).

6-Methyl-1,4-diphenylpyrimidin-2(1*H*)-one (5) reacted

with MeMgI to give only 6,6-dimethyl-1,4-diphenyl-3,6-dihydropyrimidin-2(1*H*)-one (18a) in 67% yield, while 1,6-diphenyl-4-methylpyrimidin-2(1*H*)-thione (6) with MeMgI gave only 4,4-dimethyl-1,6-diphenyl-3,4-dihydropyrimidin-2(1*H*)-thione (19b) in 82% yield (see Table).

Finally, we examined the reaction of MeMgI or MeLi with 1-phenylpyrimidin-2(1*H*)-one (7), which has no substituent at the 4- and 6-positions of the pyrimidine ring. Pyrimidin-2(1*H*)-one (7) reacted with MeMgI to yield only 6-methyl-1-phenyl-3,6-dihydropyrimidin-2(1*H*)-one (20a) in 59% yield. On the other hand, compound (7) also reacted with MeLi to give exclusively 4-methyl-1-phenyl-3,4-dihydropyrimidin-2(1*H*)-one (20b) in 88% yield (see Table).



Compound	R^4	Products	R^4MgI Yield (%)	R^4MgI Ratio (a) : (b)	R^4Li Yield (%)	R^4Li Ratio (a) : (b)
(1)	Me	(8a) and (8b)	29	95 : 5	65	15 : 85
(1)	Et	(9a) and (9b)	20	50 : 50	42	10 : 90
(1)	Pr^i	(10b)	19	0 : 100	12	0 : 100
(1)	Bu^t [a]	(11a) and (11b)	59	20 : 80		
(2)	Me	(12a) and (12b)	62	70 : 30	43	35 : 65
(2)	Et	(13a) and (13b)	83	60 : 40	42	0 : 100
(2)	Pr^i	(14a) and (14b)	48	75 : 25	10	0 : 100
(2)	Bu^t [a]	(15a) and (15b)	75	30 : 70		
(3)	Me	(16a) and (16b)	46	95 : 5	56	5 : 95
(4)	Ph–C≡C [b]	(17a)	70	100 : 0		
(5)	Me	(18a)	67	100 : 0		
(6)	Me	(19b)	82	0 : 100		
(7)	Me	(20a) and (20b)	59	100 : 0	88	0 : 100

[a] t-Butylmagnesium chloride. [b] Phenylethynylmagnesium bromide.

It is thus possible to prepare regioselectively 3,6- and 3,4-dihydropyrimidines by the reaction of pyrimidin-2(1*H*)-one and -thiones with the appropriate organometallic compounds. Further, it is concluded that alkyl Grignards attack the 6-position in preference to the 4-position of the pyrimidine ring, while alkyl-lithium reagents predominantly attack the 4-position.

EXPERIMENTAL

Materials.—Pyrimidin-2(1*H*)-ones (1) and (3) and the thione (2) were prepared by the method of Hutchins.[12] 6-Methyl-1,4-diphenylpyrimidin-2(1*H*)-one (5) and 4-methyl-1,6-diphenylpyrimidine-2(1*H*)-thione (6) were prepared from the reaction of benzolyacetone with *N*-phenylurea and *N*-phenylthiourea, respectively.[13] 1-Methylpyrimidin-2(1*H*)-one (4) [14] and 1-phenylpyrimidin-2(1*H*)-one (7) [15] were prepared by literature methods.

Reaction of Pyrimidin-2(1H)-ones and -thiones with Organometallic Compounds.—Reaction with alkyl-lithium reagents. Alkyl-lithium (10 mmol) was prepared from alkyl iodide (10 mmol) and lithium wire (25 mmol) in anhydrous ether (20 ml) uder an argon atmosphere in an ice-bath. To this solution, pyrimidin-2(1*H*)-one or -thione (3 mmol) was added and the mixture was stirred for 5 h at room temperature. Excess of alkyl-lithium was destroyed by the dropwise addition of water. The reaction mixture was extracted with dichloromethane and dried over anhydrous magnesium sulphate. The crude product was chromatographed on silica gel with chloroform–acetone–ethanol (100 : 10 : 2) in the case of pyrimidin-2(1*H*)-one, and with chloroform–benzene–ethyl acetate (4 : 4 : 1) in the case of pyrimidine-2(1*H*)-thione. In this way the following compounds were prepared: 4,6-*dimethyl*-4-*ethyl*-1-*phenyl*-3,4-*dihydropyrimidine*-2(1H)-*thione* (13b), m.p. 194—195 °C (decomp.) (from ethyl acetate); $\nu_{max.}$ (KBr) 3 180, 2 960, 1 685, 1 230, 760, and 700 cm^{-1}; δ ($CDCl_3$) 1.02 (3 H, d, *J* 6.0 Hz), 1.33 (3 H, s), 1.52 (3 H, s), 4.70 (1 H, br s), and 7.2—7.5 (5 H, m) (Found: C, 68.55; H, 7.4; N, 11.4. $C_{14}H_{18}N_2S$ requires C, 68.25; H, 7.36; N, 11.37%): 4-*isopropyl*-4,6-*dimethyl*-1-*phenyl*-3,4-*dihydropyrimidine*-2(1H)-*thione* (14b), m.p. 181—182 °C (decomp.) (from ethyl acetate–hexane); $\nu_{max.}$ (KBr) 3 180, 2 960, 1 685, 760, and 700 cm^{-1}; δ ($CDCl_3$) 0.92 (3 H, d, *J* 6.0 Hz), 1.05 (3 H, d, *J* 6.0 Hz), 1.32 (3 H, s), 1.52 (3 H, s), 4.73 (1 H, br s), and 7.2—7.5 (5 H, m) (Found: C, 69.2; H, 7.8; N, 10.7. $C_{15}H_{20}N_2S$ requires C, 69.18; H, 7.74; N, 10.75%): 4-*methyl*-1-*phenyl*-3,4-*dihydropyrimidin*-2(1H)-*one* (20b), m.p. 123—124 °C (from benzene–hexane); $\nu_{max.}$ (KBr) 3 200, 2 960, 1 690, 1 660, 760, and 690 cm^{-1}; δ ($CDCl_3$) 1.25 (3 H, d, *J* 6.0 Hz), 4.1—4.4 (1 H, m), 4.6—5.0 (1 H, m), 5.92 (1 H, br s), 6.1—6.3 (1 H, dd, *J* 8.0 and 1.0 Hz), and 7.2—7.4 (5 H, m) (Found: C, 70.5; H, 6.3; N, 14.9. $C_{11}H_{12}N_2O$ requires C, 70.19; H, 6.42; N, 14.88%).

Reaction with alkyl Grignards. Pyrimidin-2(1*H*)-one or -thione (1 mmol) was added to the solution of methylmagnesium iodide (10 mmol) in anhydrous ether (20 ml) and the mixture was stirred for 6 h. The reaction mixture was washed with cold water, extracted with dichloromethane was washed with cold water, extracted with dichloromethane, and dried over anhydrous magnesium sulphate. The crude product was chromatographed on silica gel with chloroform–acetone–ethanol (100 : 20 : 4) [(100 : 10 : 2) in

the case of pyrimidin-2(1*H*)-ones], and with chloroform–benzene–ethyl acetate (4 : 4 : 1) [(4 : 10 : 1) in the case of pyrimidin-2(1*H*)-thiones]. In this way the following compounds were prepared: *4,6,6-trimethyl-1-phenyl-3,6-dihydropyrimidin-2(1H)-one* (8a), m.p. 191—192 °C (from benzene–hexane); $\nu_{max.}$ (KBr) 3 200, 2 960, 1 660, and 760 cm^{-1}; $\lambda_{max.}$ (EtOH) 233 nm (ε 4.52 × 10^3); δ ($CDCl_3$) 1.18 (6 H, s), 170 (3 H, d, *J* 0.6 Hz), 4.45 (1 H, br s), and 7.2—7.5 (5 H, m) (Found: C, 72.2; H, 7.4; N, 13.0. $C_{13}H_{16}N_2O$ requires C, 72.19; H, 7.45; N, 12.95%): *4,4,6-trimethyl-1-phenyl-3,4-dihydropyrimidin-2(1H)-one* (8b), m.p. 163 °C (lit.,[1] 162—164 °C) (from ethyl acetate–hexane); $\nu_{max.}$ (KBr) 3 220, 2 960, 1 660, 1 395, 1 380, 760, and 695 cm^{-1}; $\lambda_{max.}$ (EtOH) 229 nm (ε 6.77 × 10^3); δ ($CDCl_3$) 1.30 (6 H, s), 1.53 (3 H, d, *J* 0.6 Hz), 4.62 (1 H, br s), 5.02 (1 H, br s), and 7.1—7.4 (5 H, m) (Found: C, 72.0; H, 7.4; N, 13.1. $C_{13}H_{16}N_2O$ requires C, 72.19; H, 7.45; N, 12.95%): *6-ethyl-4,6-dimethyl-1-phenyl-3,6-dihydropyrimidin-2(1H)-one* (9a), m.p. 143—144 °C (from benzene–hexane); $\nu_{max.}$ (KBr) 3 220, 2 960, 1 660, and 755 cm^{-1}; δ ($CDCl_3$) 1.08 (3 H, t, *J* 6.0 Hz), 1.33 (3 H, s), 1.73 (3 H, s), 4.25 (1 H, br s), and 7.0—7.2 (5 H, m) (Found: C, 72.9; H, 7.7; N, 12.2. $C_{14}H_{18}N_2O$ requires C, 73.01; H, 7.87; N, 12.16%): *4-ethyl-4,6-dimethyl-1-phenyl-3,4-dihydropyrimidin-2(1H)-one* (9b), m.p. 168 °C (from benzene–hexane); $\nu_{max.}$ (KBr) 3 230, 2 960, 1 660, and 755 cm^{-1}; δ ($CDCl_3$) 1.00 (3 H, t, *J* 6.0 Hz), 1.28 (3 H, s), 1.53 (3 H, s), 4.48 (1 H, br s), 4.76 (1 H, br s), and 7.0—7.2 (5 H, m) (Found: C, 72.5; H, 7.9; N, 12.0. $C_{14}H_{18}N_2O$ requires C, 73.01; H, 7.87; N, 12.16%): *4-isopropyl-4,6-dimethyl-1-phenyl-3,4-dihydropyrimidin-2(1H)-one* (10b), m.p. 202 °C (from benzene–hexane); $\nu_{max.}$ (KBr) 3 200, 2 960, 1 690, 1 660, 760, and 690 cm^{-1}; δ ($CDCl_3$) 0.97 (6 H, d, *J* 6.0 Hz), 1.27 (3 H, s), 1.52 (3 H, s), 4.50 (1 H, br s), 4.83 (1 H, br s), and 7.0—7.2 (5 H, m) (Found: C, 73.55; H, 8.3; N, 11.5. $C_{15}H_{20}N_2O$ requires C, 73.73; H, 8.25; N, 11.46%): *6-t-butyl-4,6-dimethyl-1-phenyl-3,6-dihydropyrimidin-1(1H)*-one (11a), m.p. 191 °C (from benzene–hexane); $\nu_{max.}$ (KBr) 3 200, 2 960, 1 700, 1 650, 1 380, and 760 cm^{-1}; δ ($CDCl_3$) 0.98 (9 H, s), 1.17 (3 H, s), 1.72 (3 H, s), 4.37 (1 H, br s), and 6.9—7.3 (5 H, m) (Found: C, 74.2; H, 8.6; N, 10.7. $C_{16}H_{22}N_2O$ requires C, 74.38; H, 8.58; N, 10.84%): *4-t-butyl-4,6-dimethyl-1-phenyl-1,2,3,4-tetrahydropyrimidine* (11b), m.p. 139 °C (from hexane); $\nu_{max.}$ (KBr) 3 240, 2 960, 1 690, 1 660, 760, and 680 cm^{-1}; δ ($CDCl_3$) 0.96 (9 H, s), 1.23 (3 H, s), 1.52 (3 H, s), 4.60 (1 H, br s), 5.07 (1 H, br s), and 7.0—7.3 (5 H, m) (Found: C, 74.6; H, 8.65; N, 10.95. $C_{16}H_{22}N_2O$ requires C, 74.38; H, 8.58; N, 10.84%): *4,6,6-trimethyl-1-phenyl-3,6-dihydropyrimidine-2(1H)-thione* (12a),* m.p. 215—223 °C (from ethyl acetate); δ ($CDCl_3$) 1.22 (6 H, s), 1.78 (3 H, d, *J* 0.6 Hz), 4.50 (1 H, br s), and 7.1—7.5 (5 H, m) {Found [for a mixture of (12a) and (12b)]: C, 67.15; H, 6.95; N, 11.95. $C_{13}H_{16}N_2S$ requires C, 67.20; H, 6.94; N, 12.05%}: *6-ethyl-4,6-dimethyl-1-phenyl-3,6-dihydropyrimidine-2(1H)-thione* (13a),* m.p. 170—179 °C (decomp.) (from ethyl acetate); δ ($CDCl_3$) 1.02 (3 H, t, *J* 6.0 Hz), 1.13 (3 H, s), 1.85 (3 H, d, *J* 0.6 Hz), 4.50 (1 H, br s), and 7.2—7.5 (5 H, m) {Found [for a mixture of (13a) and (13b)]: C, 63.25; H, 7.35; N, 11.3. $C_{14}H_{18}N_2S$ requires C, 68.25; H, 7.36; N, 11.37%}: *6-isopropyl-4,6-dimethyl-1-phenyl-3,6-dihydropyrimidine-2(1H)-thione* (14a),* m.p. 197—203 °C (decomp.) (from benzene–hexane); δ ($CDCl_3$) 0.80 (6 H, d, *J* 6.0 Hz), 1.15 (3 H, s), 1.83 (3 H, s), 4.48 (1 H, br s), 7.2—7.5 (5 H, m), and 8.27 (1 H, br s) {Found [for a mixture of (14a) and (14b)]: C, 69.25; H, 7.8; N, 10.7. $C_{15}H_{20}N_2O$ requires C, 69.18; H, 7.74; N, 10.75%}: *6-t-butyl-4,6-dimethyl-1-phenyl-3,6-dihydropyrimidine-2(1H)-thione* (15a), m.p. 167—168 °C (decomp.) (from ethyl acetate–hexane); $\nu_{max.}$ (KBr) 3 240, 2 960, 1 710, 1 240, and 710 cm^{-1}; δ ($CDCl_3$) 1.03 (9 H, s), 1.18 (3 H, s), 1.85 (3 H, s), 4.60 (1 H, br s), 7.1—7.5 (5 H, m), and 8.43 (1 H, br s) (Found: C, 69.8; H, 7.9; N, 10.1. $C_{16}H_{22}N_2S$ requires C, 70.02; H, 8.08; N, 10.20%): *4-t-butyl-4,6-dimethyl-1-phenyl-3,4-dihydropyrimidine-2(1H)-thione* (15b), m.p. 180 °C (decomp.) (from benzene–hexane); $\nu_{max.}$ (KBr) 3 180, 2 960, 1 680, 1 600, and 760 cm^{-1}; δ ($CDCl_3$) 1.01 (9 H, s), 1.28 (3 H, s), 1.53 (3 H, s), 4.75 (1 H, br s), and 7.1—7.4 (5 H, m) (Found: C, 70.0; H, 8.0; N, 10.2. $C_{16}H_{22}N_2S$ requires C, 70.02; H, 8.08; N, 10.20%): *4,6,6-trimethyl-1-p-tolyl-3,6-dihydropyrimidine-2(1H)-one* (16a), m.p. 211—212 °C (from ethyl acetate–hexane); $\nu_{max.}$ (KBr) 3 200, 2 960, 1 710, 1 660, 1 400, 810, and 740 cm^{-1}; δ ($CDCl_3$) 1.20 (6 H, s), 1.75 (3 H, d, *J* 0.6 Hz), 2.36 (3 H, s), 4.47 (1 H, br s), and 7.1—7.2 (4 H, m) (Found: C, 73.6; H, 7.95; N, 12.15. $C_{14}H_{18}N_2O$ requires C, 73.01; H, 7.87; N, 12.16%): *4,4,6-trimethyl-1-p-tolyl-3,4-dihydropyrimidin-2(1H)-one* (16b), m.p. 193 °C (from ethyl acetate–hexane); $\nu_{max.}$ (KBr) 3 220, 2 960, 1 700, 1 670, 1 510, 805, and 750 cm^{-1}; δ ($CDCl_3$) 1.32 (6 H, s), 1.51 (3 H, d, *J* 0.6 Hz), 2.35 (3 H, s), 4.64 (1 H, br s), 5.04 (1 H, br s), and 7.1—7.2 (4 H, m) (Found: C, 73.3; H, 7.9; N, 12.5. $C_{14}H_{18}N_2O$ requires C, 73.01; H, 7.87; N, 12.16%): *1-methyl-6-phenylethynyl-3,6-dihydropyrimidin-2(1H)-one* (17a), m.p. 134—135 °C (from ethyl acetate–hexane); $\nu_{max.}$ (KBr) 3 220, 2 940, 1 680, and 750 cm^{-1}; δ ($CDCl_3$) 3.04 (3 H, s), 4.6—5.0 (2 H, m), 6.0—6.3 (1 H, dd, *J* 7.0 and 4.8 Hz), 7.2—7.5 (5 H, m), and 8.50 (1 H, br s) (Found: C, 73.7; H, 5.65; N, 13.2. $C_{13}H_{12}N_2O$ requires C, 73.56; H, 5.69; N, 13.19%): *6,6-dimethyl-1,4-diphenyl-3,6-dihydropyrimidin-2(1H)-one* (18a), m.p. 196—197 °C (from ethyl acetate); $\nu_{max.}$ (KBr) 3 220, 2 980, 1 640, 1 410, 760, and 740 cm^{-1}; δ ($CDCl_3$) 1.33 (6 H, s), 4.93 (1 H, br s), and 7.0—7.5 (10 H, m) (Found: C, 77.7; H, 6.45; N, 10.05. $C_{18}H_{18}N_2O$ requires C, 77.66; H, 6.51; N, 10.06%): *4,4-dimethyl-1,6-diphenyl-3,4-dihydropyrimidine-2(1H)-thione* (19b), m.p. 206—207 °C (decomp.) (from benzene–hexane); $\nu_{max.}$ (KBr) 3 200, 2 980, 1 680, 1 540, 1 340, 1 200, and 700 cm^{-1}; δ ($CDCl_3$) 1.48 (6 H, s), 5.10 (1 H, br s), and 7.0—7.3 (10 H, m) (Found: C, 73.5; H, 6.10; N, 9.55. $C_{18}H_{18}N_2S$ requires C, 73.43; H, 6.16; N, 9.51%): *6-methyl-1-phenyl-3,6-dihydropyrimidin-2(1H)*-one (20a), m.p. 140.5 °C (decomp.) (from ethyl acetate–hexane); $\nu_{max.}$ (KBr) 3 220, 2 960, 1 650, 1 190, and 760 cm^{-1}; δ ($CDCl_3$) 1.17 (3 H, d, *J* 6.0 Hz), 4.2—4.9 (2 H, m), 5.9—6.2 (1 H, dd, *J* 8.0 and 5.6 Hz), 7.2—7.5 (5 H, m), and 8.35 (1 H, br s) (Found: C, 69.95; H, 6.4; N, 14.8. $C_{11}H_{12}N_2O$ requires C, 70.19; H, 6.42; N, 14.88%).

* Attempts to separate the mixture by column chromatography or fractional recrystallization was unsuccessful.

[0/787 *Received*, 23*rd May*, 1980]

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