Synthesis of 2,3-Dihydro-2-alkyl-3-(substituted amino)-1*H*-isoindol-1-ones. 2-Alkylation of 2,3-Dihydro-3-(substituted amino)-1*H*-isoindol-1-ones in a Phase-Transfer Catalyst System

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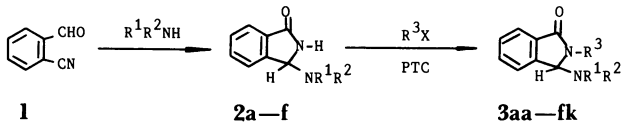
Synopsis. A variety of 2,3-dihydro-2-alkyl-3-(substituted amino)-1*H*-isoindol-1-ones were conveniently synthesized by selective 2-alkylation of 2,3-dihydro-3-(substituted amino)-1*H*-isoindol-1-ones, which were derived from 2-cyanobenzaldehyde, with some alkyl halides in a phase-transfer catalyst system.

We recently reported the formation of γ-lactams, 2,3-dihydro-3-(substituted amino)-1*H*-isoindol-1-ones, from the reaction of 2-cyanobenzaldehyde with primary and secondary alkylamines at room temperature in excellent yields, and also revealed that the amines used were introduced regioselectively into the 3-position, not into the 2-position, of the 2,3-dihydro-3-(substituted amino)-1*H*-isoindol-1-ones.[1] Since 2-alkylated isoindol-1-ones could not be obtained directly by the method, our interests focused on the selective alkylation at the 2-position of 2,3-dihydro-3-(substituted amino)-1*H*-isoindol-1-ones. Although, to our knowledge, there have been no reports on the direct 2-alkylation of the isoindol-1-ones, the selective *N*-alkylation of amides in the presence of a phase-transfer catalyst have recently appeared.[2] We also reported the one-pot synthesis of alkyl aryl trithiocarbonates from benzenethiols and carbon disulfide with alkyl halides in the presence of a phase-transfer catalyst in good yields.[3] Thus, because of the experimetal simplicity, superior regioselectivity and high yields of the products, the phase-transfer catalyst(PTC) has been used widely for alkylations of some nitrogen or sulfur organic compounds which are difficult to be alkylated regioselectively.[4] This elegant technique encouraged us to explore the regioselective 2-alkylation of 2,3-dihydro-3-(substituted amino)-1*H*-isoindol-1-ones with alkyl halides. In this paper, we wish to report a first example for the 2-alkylation of 2,3-dihydro-3-(substituted amino)-1*H*-isoindol-1-ones(**2**), which are derived from 2-cyanobenzaldehyde(**1**),[1] in the phase-transfer catalyst(PTC) system to give 2,3-dihydro-2-alkyl-3-(substituted amino)-1*H*-isoindol-1-ones(**3**) in high yields (Scheme 1).

Table 1. 2-Alkylations of 2,3-Dihydro-3-(substituted amino)-1*H*-isoindol-1-ones (**3**) with Alkyl halides in a Phase-Transfer Catalyst System

Run[a]	Substrate		Halide		React		Yield of **3**		Mp
	R^1	R^2		R^3X	Temp/°C	Time/min	% [b]		θ_m/°C
1	H	H	**2a**	$4\text{-}CH_3C_6H_4CH_2Br$	20	10	75	**3aa**	106
2			**2a**	$BrCH_2COOC_2H_5$	20	20	90	**3ab**	77
3	H	CH_3	**2b**	$2\text{-}ClC_6H_4CH_2Cl$	20	3(h)	83	**3bc**	107
4			**2b**	$3,4\text{-}Cl_2C_6H_3CH_2Cl$	20	3(h)	66	**3bd**	108
5	H	$C(CH_3)_3$	**2c**	CH_3I	20	5(h)	83	**3ce**	171
6			**2c**	$C_6H_5CH{=}CHCH_2Br$	20	60	90	**3cf**	137
7			**2c**	$2,4\text{-}Cl_2C_6H_3CH_2Cl$	20	2(h)	63	**3cg**	157
8	C_2H_5	C_2H_5	**2d**	CH_3I	20	24(h)	94	**3de**	53
9			**2d**	$ClCH_2CN$	20	30	95	**3dh**	69
10			**2d**	$C_6H_5CH_2Br$	20	10	94	**3di**	79
11			**2d**	$4\text{-}CH_3C_6H_4CH_2Br$	20	10	97	**3da**	64
12			**2d**	$2\text{-}CH_3C_6H_4CH_2Br$	60	10	96	**3dj**	87
13			**2d**	$4\text{-}ClC_6H_4CH_2Cl$	60	90	95	**3dk**	69
14			**2d**	$2\text{-}ClC_6H_4CH_2Cl$	60	40	94	**3dc**	88
15			**2d**	$2,4\text{-}Cl_2C_6H_3CH_2Cl$	60	30	91	**3dg**	126
16			**2d**	$3,4\text{-}Cl_2C_6H_3CH_2Cl$	60	15	92	**3dd**	74
17			**2d**	$CH{\equiv}CCH_2Br$	60	4(h)	90	**3dl**	62
18	$-CH_2CH_2OCH_2CH_2-$		**2e**[6]	CH_3I	20	4(h)	92	**3ee**	115
19			**2e**	$CH_2{=}CHCH_2Br$	20	4(h)	97	**3em**	64
20			**2e**	$2\text{-}CH_3C_6H_4CH_2Br$	20	10	97	**3ej**	114
21	$-CH_2(CH_2)_3CH_2-$		**2f**[6]	$ClCH_2CN$	20	40	54	**3fh**	98
22			**2f**	$4\text{-}ClC_6H_4CH_2Cl$	20	2(h)	91	**3fk**	121

a) Tetrabutylammonium iodide was used as phase-transfer catalyst. b) Isolated yields based on the substrates.



1 2a—f 3aa—fk

Scheme 1.

Many 2,3-dihydro-2-alkyl-3-(substituted amino)-1*H*-isoindol-1-ones(**3**) were obatained in high yields as shown in Table 1. It should be noted that the synthesis of many functionalized isoindol-1-ones(**3**) containing cyano, chloro and unsaturated alkyl groups in the molecules as shown in the table was carried out successfully, of which antiflammatory activity has been observed.[5] In the absence of the phase-transfer catalyst, the substrate, isoindol-1-one(**2**), was not alkylated at all. Various quarternary ammonium salts, for example, methyltrioctylammonium chloride(TOMAC) and tetrabutylammonium bromide and iodide could be used, and little difference by the catalyst was observed in the present reaction. On the other hand, a considerable increase of the yield of 2-alkylated isoindol-1-ones(**3**) could be confirmed when the concentration of sodium hydroxide aqueous solution was changed to 50% from 20%. It is noteworthy that a high selectivity was observed in this reaction. Thus, 2-alkylated isoindol-1-ones(**3aa-cg**) were obtained by treating 3-amino(**2a**), 3-methylamino(**2b**), and 3-(*t*-butylamino)-2,3-dihydro-1*H*-isoindol-1-one(**2c**) with alkyl halide in the presence of tetrabutylammonium iodide in high yields without any 3-(alkylated amino)-2,3-dihydro-1*H*-isoindol-1-one (Runs 1—7). These reactions proceeded smoothly at 20 °C to 60 °C.

Consequently, a novel route to 2-alkyl-3-(substituted amino)-2,3-dihydro-1*H*-isoindol-1-ones(**3**) from 2-cyanobenzaldehyde(**1**) was confirmed.

Experimental

Measurements. All the melting points were uncorrected. NMR, IR and Mass spectra were recorded with a HITACHI R-22, HITACHI 295 and HITACHI RMU-6M, respectively. Elemental analysis were determined with YANAGIMOTO MT-3.

Materials. 2,3-Dihydro-3-(substituted amino)-1*H*-isoindol-1-ones(**2**) were obtained from reactions of 2-cyanobenzaldehyde(**1**) with the corresponding amines.[1]

General Procedure for the Preparation of 2,3-Dihydro-2-alkyl-3-(substituted amino)-1*H*-isoindol-1-ones(3) from 2,3-Dihydro-3-(substituted amino)-1*H*-isoindol-1-ones(2). An alkyl halide (1.1 mmol) was added dropwise to a mixture of 1.0 mmol of 2,3-dihydro-3-(substituted amino)-1*H*-isoindol-1-ones(**2**), 0.2 mmol of tetrabutylammonium iodide, 5 ml of benzene and 5 ml of 50% aqueous sodium hydroxide. The order of addition of the materials did not affect the results. After the completion of the addition, stirring was continued at 20 °C until the substrate disappeared (monitoring by TLC). Water (5 ml) was added to the reaction mixture and then the benzene layer was separated. The aqueous layer was extracted with benzene three times (5 ml each), and the organic layers were combined. Evaporation of the solvent from the combined benzene solution after drying with magnesium sulfate gives a yellow oil. Desired 2,3-dihydro-2-alkyl-3-(substituted amino)-1*H*-isoindol-1-ones(**3**) were obtained as clear colorless crystals by column chromatgraphy of the oil on silica gel (Wako gel C-300) using chloroform as eluent, followed by repeated recrytallization from chloroform–hexane. All products were new compounds, being assigned by physical, spectral and analytical data as follows.

3aa. Colorless crystals, mp 106 °C; IR (KBr) 3370, 3000, 2900, and 1685 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.80 (s, 2H, NH_2), 2.31 (s, 3H, CH_3), 4.37 and 5.10 (d, 2H, J=15.0 Hz, CH_2), 5.90 (s, 1H, CH) and 7.00—7.90 (m, 8H, arom); MS (70 eV) *m/z* 252 (M^+); Found: C, 76.14; H, 6.39; N, 11.10%. Calcd for $C_{16}H_{16}N_2O$: C, 76.16; H, 6.39; N, 11.10%.

3ab. Colorless crystals, mp 77 °C; IR (KBr) 3390, 3300, 2960, 1760, and 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.27 (t, 3H, 6.5 Hz, CH_3), 1.84 (s, 2H, NH_2), 4.14 and 4.52 (d, 2H, 17.0 Hz, CH_2), 4.18 (d, 2H, 6.5 Hz, CH_2), 5.42 (s, 1H, CH) and 7.45—7.85 (m, 4H, arom); MS (70 eV) *m/z* 234 (M^+); Found: C, 61.32; H, 6.02; N, 11.67%. Calcd for $C_{12}H_{14}N_2O_3$; C, 61.53; H, 6.02; N, 11.96%.

3bc. Colorless crystals, mp 107 °C; IR (KBr) 3340, 2925, and 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.94 (s, 3H, CH_3), 2.08 (bs, 1H, NH), 4.50 and 5.16 (d, 2H, 15.0 Hz, CH_2), 5.27 (s, 1H, CH) and 7.10—7.90 (m, 8H, arom); MS (70 eV) *m/z* 287 (M^+); Found: C, 66.88; H, 5.18; N, 9.76%. Calcd for $C_{16}H_{15}N_2OCl$: C, 67.02; H, 5.27; N, 9.77%.

3bd. Colorless crystals, mp 108 °C; IR (KBr) 3320, 2900, and 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ=2.00 (s, 3H, CH_3), 4.28 and 5.09 (d, 2H, 15.0 Hz, CH_2), 5.22 (s, 1H, CH), 7.08—7.92 (m, 7H, arom); MS (70 eV) *m/z* 321 (M^+); Found: C, 59.65; H, 4.32; N, 8.72%. Calcd for $C_{16}H_{14}N_2OCl_2$: C, 59.83; H, 4.39; N, 8.72%.

3ce. Colorless crystals, mp 171 °C; IR (KBr) 3325, 2955, and 1670 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.33 (s, 9H, CH_3), 3.07 (s, 3H, CH_3), 5.20 (s, 1H, CH) and 7.35—7.83 (m, 4H, arom); MS (70 eV) *m/z* 218 (M^+); Found: C, 71.12; H, 8.43; N, 12.87%. Calcd for $C_{13}H_{18}N_2O$: C, 71.53; H, 8.31; N, 12.83%.

3cf. Colorless crystals, mp 137 °C; IR (KBr) 3320, 2960, 2900, and 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.30 (s, 9H, CH_3), 1.40 (b, 1H, NH), 4.07 (dd, 1H, 16.0,6.5 Hz, CH_2), 4.64 (dd, 1H, 16.0, 3.5 Hz, CH_2), 5.35 (s, 1H, CH), 6.00—6.60 (m, 2H, CH=CH) and 7.10—7.88 (m, 9H, arom); MS (70 eV) *m/z* 320 (M^+); Found: C, 78.67; H, 7.64; N, 8.84%. Calcd for $C_{21}H_{24}N_2O$: C, 78.71; H, 7.55; N, 8.74%.

3cg. Colorless crystals, mp 157 °C; IR (KBr) 3350, 2970, and 1700 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.16 (s, 9H, CH_3), 1,47 (b, 1H, NH), 4.85 (d, 2H, 6.0 Hz, CH_2), 5.28 (s, 1H, CH) and 6.89—7.90 (m, 7H, arom); MS (70 eV) *m/z* 363 (M^+); Found: C, 62.76; H, 5.49; N, 7.69%. Calcd for $C_{19}H_{20}N_2OCl_2$: C, 62.82; H, 5.55; N, 7.71%.

3de. Colorless crystals, mp 53 °C; IR (KBr) 2960, 2825, and 1700 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.06 (t, 6H, 7.5 Hz, CH_3), 2.62 (q, 4H, 7.5 Hz, CH_2), 3.08 (s, 3H, CH_3), 5.22 (s, 1H, CH) and 7.30—7.86 (m, 4H, arom); MS (70 eV) *m/z* 218 (M^+); Found: C, 71.29; H, 8.23; N, 12.60%. Calcd for $C_{13}H_{18}N_2O$: C, 71.53; H, 8.31; N, 12.83%.

3dh. Colorless crystals, mp 69 °C; IR (KBr) 2975, 2840, 2250, and 1710 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.13 (t, 6H, 7.5 Hz, CH_3), 2.64 (q, 4H, 7.5 Hz, CH_2), 4.22 and 4.75 (d, 2H, 17.5 Hz, CH_2), 5.48 (s, 1H, CH) and 7.35—7.93 (m, 4H, arom); MS (70 eV) *m/z* 243 (M^+); Found: C, 69.00; H, 7.09; N, 16.98%. Calcd for $C_{14}H_{17}N_3O$: C, 69.11; H, 7.04; N, 17.27%.

3di. Colorless crystals, mp 79 °C; IR (KBr) 2950, 2820, and 1680 cm^{-1}; ^1H NMR ($CDCl_3$) δ=0.98 (t, 6H, 7.3 Hz, CH_3), 2.54 (q, 4H, 7.3 Hz, CH_2), 4.24 and 5.23 (d, 2H, 15.0 Hz, CH_2), 5.15 (s, 1H, CH) and 7.20—7.95 (m, 9H, arom); MS (70 eV) *m/z* 294 (M^+); Found: C, 77.22; H, 7.54; N, 9.31%. Calcd for $C_{19}H_{22}N_2O$: C, 77.52; H, 7.53; N, 9.52%.

3da. Colorless crystals, mp 64 °C; IR (KBr) 2960, 2825, and 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.00 (t, 6H, 7.5 Hz, CH_3), 2.30 (s, 3H, CH_3), 2.54 (q, 4H, 7.5 Hz, CH_2), 4.19 and 5.20 (d, 2H, 15.0 Hz, CH_2), 5.14 (s, 1H, CH) and 6.98—7.88 (m, 8H, arom); MS (70 eV) *m/z* 308 (M^+); Found: C, 77.78; H, 7.97; N, 8.93%. Calcd for $C_{20}H_{24}N_2O$: C, 77.89; H, 7.84; N, 9.08%.

3dj. Colorless crystals, mp 87 °C; IR (KBr) 2960, 2840, and 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ=0.98 (t, 6H, 7.3 Hz, CH_3), 2.37 (s, 3H, CH_3), 2.53 (q, 4H, 7.3 Hz, CH_2), 4.30 and 5.20 (d, 2H, 15.0 Hz, CH_2), 5.10 (s, 1H, CH) and 7.08-7.95 (m, 8H, arom); MS (70 eV) *m/z* 308 (M^+); Found: C, 77.85; H, 7.93; N, 9.00%. Calcd for $C_{20}H_{24}N_2O$: C, 77.89; H, 7.84; N, 9.08%.

3dk. Colorless crystals, mp 69 °C; IR (KBr) 2950, 2820, and 1680 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.00 (t, 6H, 7.5 Hz, CH_3), 2.54 (q, 4H, 7.5 Hz, CH_2), 4.26 and 5.13 (d, 2H, 15.0 Hz, CH_2), 5.14 (s, 1H, CH), 7.25 (s, 4H, arom) and 7.35—7.93 (m, 4H, arom); MS (70 eV) *m/z* 329 (M^+); Found: C, 69.01; H, 6.39; N, 8.54%. Calcd for $C_{19}H_{21}N_2OCl$: C, 69.40; H, 6.44; N, 8.51%.

3dc. Colorless crystals, mp 88 °C; IR (KBr) 2960, 2830, and 1690 cm^{-1}; ^1H NMR ($CDCl_3$) δ=0.97 (t, 6H, 7.5 Hz, CH_3), 2.53 (q, 4H, 7,5 Hz, CH_2), 4.63 and 5.13 (d, 2H, 15.5 Hz, CH_2), 5.24 (s, 1H, CH) and 7.10—7.96 (m, 8H, arom); MS (70 eV) *m/z* 329 (M^+); Found: C, 69.17; H, 6.44; N, 8.47%. Calcd for $C_{19}H_{21}N_2OCl$: C, 69.40; H, 6.44; N, 8.51%.

3dg. Colorless crystals, mp 126 °C; IR (KBr) 2960, 2825, and 1700 cm^{-1}; ^1H NMR ($CDCl_3$) δ=0.97 (t, 6H, 7.4 Hz, CH_3), 2.52 (q, 4H, 7.4 Hz, CH_2), 4.60 and 5.04 (d, 2H, 15.5 Hz, CH_2), 5.20 (s, 1H, CH) and 7.10—7.93 (m, 7H, arom); MS (70 eV) *m/z* 363 (M^+); Found: C, 62.58; H, 5.41; N, 7.64%. Calcd for $C_{19}H_{20}N_2OCl_2$: C, 62.82; H, 5.55; N, 7.71%.

3dd. Colorless crystals, mp 74 °C; IR (KBr) 2960, 2825, and 1690 cm^{-1}: ^1H NMR ($CDCl_3$) δ=1.00 (t, 6H, 7.4 Hz, CH_3), 2.53 (q, 4H, 7.4Hz, CH_2), 4.24 and 5.07 (d, 2H, 15.0 Hz, CH_2), 5.13 (s, 1H, CH) and 7.05—7.94 (m, 7H, arom); MS (70 eV) *m/z* 363 (M^+); Found: C, 62.20; H, 5.57; N, 7.62%. Calcd for $C_{19}H_{20}N_2OCl_2$: C, 62.82; H, 5.55; N, 7.71%.

3dl. Colorless crystals, mp 62 °C; IR (KBr) 3230, 2960, 2800, and 1700 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.07 (t, 6H, 7.5 Hz, CH_3), 2.18—2.27 (m, 1H, -C≡CH), 2.62 (q, 4H, 7.5 Hz, CH_2), 3.94, and 4.74 (dd, 2H, 17.0, 2.8 Hz, CH_2), 5.51 (s, 1H, CH) and 7.25—7.87 (m, 4H, arom); MS (70 eV) *m/z* 242 (M^+); Found: C, 74.14; H, 7.44; N, 11.44%. Calcd for $C_{15}H_{18}N_2O$: C, 74.35; H, 7.49; N, 11.56%.

3ee. Colorless crystals, mp 115°C; IR (KBr) 2870, 2820, and 1700 cm^{-1}; ^1H NMR ($CDCl_3$) δ=2.40—2.80 (m, 4H, CH_2), 3.14 (s, 3H, CH_3), 3.67 (t, 4H, 4.8 Hz, CH_2), 5.04 (s, 1H, CH) and 7.36—7.88 (m, 4H, arom); MS (70 eV) *m/z* 232 (M^+); Found: C, 67.20; H, 7.01; N, 11.76%. Calcd for $C_{13}H_{16}N_2O_2$: C, 67.22; H, 6.94; N, 12.06%.

3em. Colorless crystals, mp 64 °C; IR (KBr) 2945, 2840, and 1700 cm^{-1}; ^1H NMR ($CDCl_3$) δ=2.35—2.83 (m, 4H, CH_2), 3.65 (t, 4H, 4.8 Hz, CH_2), 3.87 (dd, 1H, 15.0, 7.3 Hz, CH_2), 4.53 (dd, 1H, 15.0, 5.0 Hz, CH_2), 5.10—5.33 (m, 2H, -C=CH_2), 5.14 (s, 1H, CH), 5.63—6.06 (m, 1H, -CH=C) and 7.30—7.90 (m, 4H, arom); MS (70 eV) *m/z* 258 (M^+); Found: C, 69.68; H, 7.11; N, 11.79%. Calcd for $C_{15}H_{18}N_2O_2$: C, 69. 74; H, 7.02; N, 10.84%.

3ej. Colorless crystals, mp 114 °C; IR (KBr) 2940, 2840, and 1700 cm^{-1}; ^1H NMR ($CDCl_3$) δ=2.24—2.84 (m, 4H, CH_2), 2.39 (s, 3H, CH_3), 3.68 (t, 4H, 4.8 Hz, CH_2), 4.34 and 5.23 (d, 2H, 14.5 Hz, CH_2), 4.86 (s, 1H, CH) and 7.10—7.98 (m, 8H, arom); MS (70 eV) *m/z* 322 (M^+); Found: C, 74.53; H, 6.94; N, 8.54%. Calcd for $C_{20}H_{22}N_2O_2$: C, 74.51; H, 6.88; N, 8.69%.

3fh. Colorless crystals, mp 98 °C; IR (KBr) 2940, 2800, and 1705 cm^{-1}; ^1H NMR ($CDCl_3$) δ=1.30—1.75 (b, 6H, CH_2), 2.30—2.75 (b, 4H, CH_2), 4.27 and 4.68 (d, 2H, 17.0 Hz, CH_2), 5.25 (s, 1H, CH) and 7.35—7.90 (m, 4H, arom); MS (70 eV) *m/z* 255 (M^+); Found: C, 70.33; H, 6.69; N, 16.51%. Calcd for $C_{15}H_{17}N_3O$: C, 70.56; H, 6.71; N, 16.46%.

3fk. Colorless crystals, mp 121 °C; IR (KBr) 2930, 2800, and 1700 cm^{-2}; ^1H NMR ($CDCl_3$) δ=1.35—1.70 (b, 6H, CH_2), 2.20—2.70 (b, 4H, CH_2), 4.28 and 5.09 (d, 2H, 14.0 Hz, CH_2), 4.89 (s, 1H, CH), 7.24 (s, 4H, arom) and 7.35—7.90 (m, 4H, arom); MS (70 eV) *m/z* 341 (M^+); Found: C, 70.42; H, 6.24; N, 8.40%. Calcd for $C_{20}H_{21}N_2OCl$: C, 70.48; H, 6.21; N, 8.22%.

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