A Convenient Preparation of Some *N*-Alkylcarbazoles and *N*-Alkylacridones

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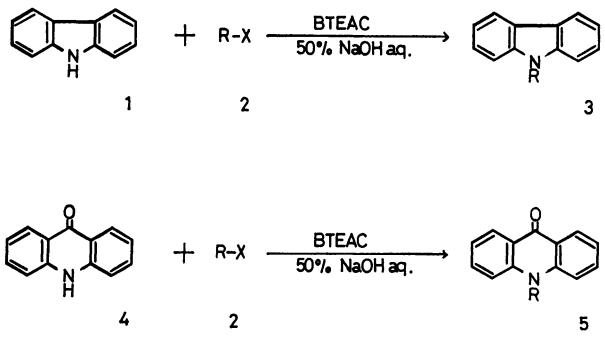
Synopsis. *N*-Alkylation of aromatic compounds involving nitrogen heterocycles such as carbazole and acridone with alkyl halide in the presence of caustic solution and benzyl triethyl ammonium chloride (BTEAC) as a phase-transfer catalyst readily proceeded under mild conditions. These results show that this procedure is effective for the preparation of the title compounds in high yields.

A few investigations of *N*-alkylation of aromatic compounds involving nitrogen heterocycles with alkyl halide under phase-transfer catalytic conditions have been reported. Makosza,[1] for example, synthesized some *N*-alkylindoles and *N*-butylcarbazole by the use of phase-transfer catalysis in more than 80% yield under mild conditions. On the other hand, Kricka *et al.*[2] studied the synthesis of *N*-alkylcarbazoles, which were prepared by the reaction of carbazole, alkyl halide, and thallium(I) ethoxide under mild conditions.

Recently, Galy *et al.*[3] have reported the reaction of acridone with alkyl halide by the use of the phase-transfer catalysis under severe conditions (refluxing in toluene for 5 d). This procedure afforded a mixture of *N*-alkylacridones (41—65%) and *O*-alkyl-acridones.



Scheme 1.

In the course of our synthetic studies of nitrogen heterocycles as intermediates of dyes and pigments, we have attempted to extend the phase-transfer catalytic reaction on *N*-alkylation of carbazole and acridone derivatives. We selectively obtained the desired *N*-alkyl compounds in high yields under milder conditions than those in Galy's procedure. *N*-Alkylation takes place as shown in Scheme 1.

The results are summarized in Table 1.

Experimental

The products were identified by the examinations of their melting points and N elemental analysis. Typical procedures are as follows:

*9-Methylcarbazole (**3a**).* To a mixture of 10.03 g (0.06 mol) of carbazole, 35 ml of aqueous 50% sodium hydroxide, 5 ml of benzene as a solvent and 410 mg (1.8 mmol) of BTEAC, a 5.6 ml (0.09 mol) of methyl iodide was added dropwise under stirring. It was continued at room temperature for 2 h. The reaction mixture was poured into hot water and left overnight at room temperature. The precipitated solid was collected, washed with water and dried. Recrystallizations from ethanol afforded 8.95 g of colorless plates (mp 88—89 °C) in the yield of 82.3%.

*10-Methylacridone (**5a**).* Methyl iodide (1.64 ml, 0.0263 mol) was added dropwise under stirring to a mixture of 3.42 g (0.0175 mol) of acridone, 13.5 ml of aqueous 50% sodium hydroxide, 13.6 ml of ethyl methyl ketone as a solvent, and 120 mg (0.527 mmol) of BTEAC, and was stirred at 55—60 °C for 3 h. The reaction mixture was poured into hot water, and separated solid worked up in the manner used for **3a**. Recrystallizations from ethanol provided 3.30 g of pale yellow needles (mp 200—201 °C) in the yield of 82.7%.

Table 1. Preparations of some *N*-alkylcarbazoles and *N*-alkylacridones

Compound No. [f,g]	RX	RX[a] / 1 or 4	Temp / °C	Time[b] / h	Products yield/% (Lit)	Mp/°C[c,d] (Lit)	Appearance
3a	CH_3I	1.50	r.t.	2	82.3(78)	88—89(87)	Colorless plates
3b	C_2H_5Br	1.50	r.t.	2	86.2(85)	67—68(67—68)	Colorless needles
3c	*n*-C_3H_7Br	1.25	55—60	1	81.0(72)	48—49(50)	Colorless needles
3d	*n*-C_4H_9Br	1.25	70—75	1	86.5(71)[e]	58—59(58)	Colorless needles
3e	$PhCH_2Cl$	1.25	70—75	1	92.0(97)	118—119(118—119)	Colorless needles
3f	$CH_2{=}CH_2CH_2Cl$	1.50	r.t.	2	74.3(67)	55—56(56)	Colorless plates
5a	CH_3I	1.50	55—60	3	82.7(45)	200—201(201)	Pale yellow needles
5b	C_2H_5Br	1.50	55—60	2	79.3(45)	158—160(158)	Yellow plates
5c	*n*-C_3H_7Br	1.50	65—70	2	75.1(43)	131—132(130)	Yellow prisms
5d	*n*-C_4H_9Br	1.50	70—75	1	80.0(65)	97—98(98)	Yellow needles
5e	$PhCH_2Cl$	1.20	70—75	1	75.7(63)	180—181(181)	Yellow needles

a) Molar ratio. b) r.t.: Room temperature. c, d) Carbazole: L. J. Kricka and A. Ledwith, *J. Chem. Soc., Perkin Trans. 1*, **1972**, 2292. Acridone: J. P. Galy and J. Barbe, *Synthesis*, **1979**, 944. e) Yield 84%, M. Makosza, *Rocz. Chem.*, **49**, 1203 (1975). f) Molar ratio of BTEAC and 1 or 4 is 0.03 : 1. g) Solvent is benzene in the case of carbazole and is ethyl methyl ketone in the case of acridone.

References

1) M. Makosza, *Rocz. Chem.*, **49**, 1203 (1975).
2) L. J. Kricka and A. Ledwith, *J. Chem. Soc., Perkin Trans. 1*, **1972**, 2292.
3) J. P. Galy and J. Barbe, *Synthesis*, **1979**, 944.