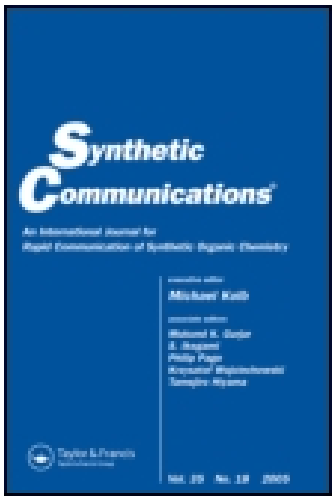

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INFLUENCE OF THE REACTION CONDITIONS IN THE N-ALKYLATION OF ACRIDONE-2-CARBOXYLIC ACIDS

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ABSTRACT

The synthesis and a study about the influence of the solvent in the alkylation reaction of 2-Carboxy-6-chloro-9(10H) acridone (3b) and 2-Carboxy-6-chloro-7-nitro-9(10H) acridone (3a) was done, obtaining the N-ethyl ester derivative of the alkylated products. Using the liquid-liquid phase transfer catalysis only the N-alkylated derivative was obtained in both cases with an improved yield.

Various N-alkylated derivatives of acridone have been described as antiallergic,[1–3] antiviral[4–8] and anticancer[9–12] compounds. As a part of our investigations in acridone derivatives we were interested in the synthesis of a new 2-Carboxy-6-chloro-7-nitro-9(10H) acridone (3a) and the corresponding N-ethyl alkylated derivative.

The alkylation of acridones has been reported in several studies,[13–15] alkylation of acridones-2-carboxylic acids using organic solvents was reported by Gorving,[2] but the alkylation of these derivatives using phase

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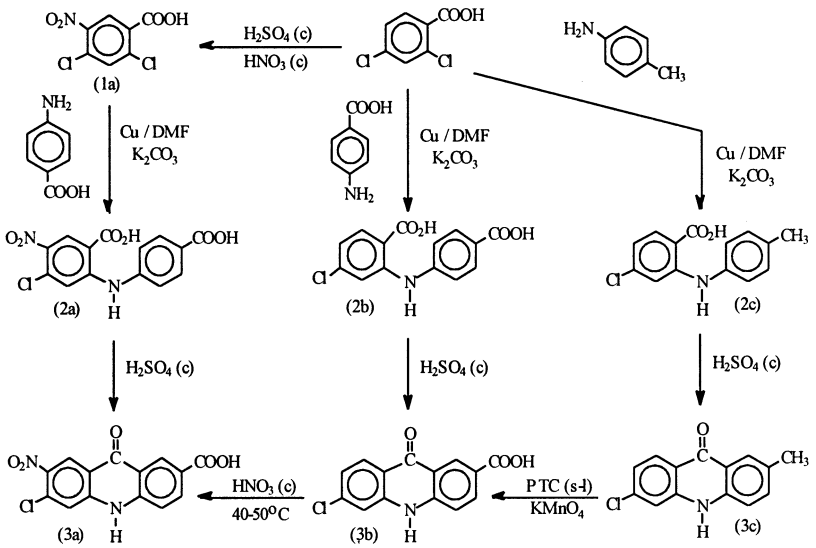
transfer catalysis conditions has not been reported. The purpose of this paper is to present the influence of the reaction conditions in the alkylation of 2-Carboxy-6-chloro-9(10H) acridone and 2-Carboxy-6-chloro-7-nitro-9(10H) acridone.

RESULTS AND DISCUSSION

The synthetic route to the obtention of 2-carboxy-6-chloro-7-nitro-9(10H) acridone is outlined in Scheme I.



Scheme I.

2,4-Dichloro-5-nitrobenzoic acid (1a) was prepared by the procedure described in literature[16] from 2,4-Dichlorobenzoic acid. The 2,4-Dichloro-5-nitro-benzoic acid (1a) obtained was condensed with p-aminobenzoic acid under conditions previously reported[17] using DMF as solvent to give the 2[(4-Carboxyphenyl)amino] 5-chloro-4-nitro benzoic acid (2a) which was cyclized to the required 2-Carboxy-6-chloro-7-nitro-9(10-H) acridone (3a) with sulphuric acid in one hour reaction time. 2-Carboxy-6-chloro-9(10-H) acridone (3b) and 2-Methyl-6-chloro-9(10-H) acridone (3c) were prepared similarly from the corresponding acids (2b) and (2c). The methyl derivative

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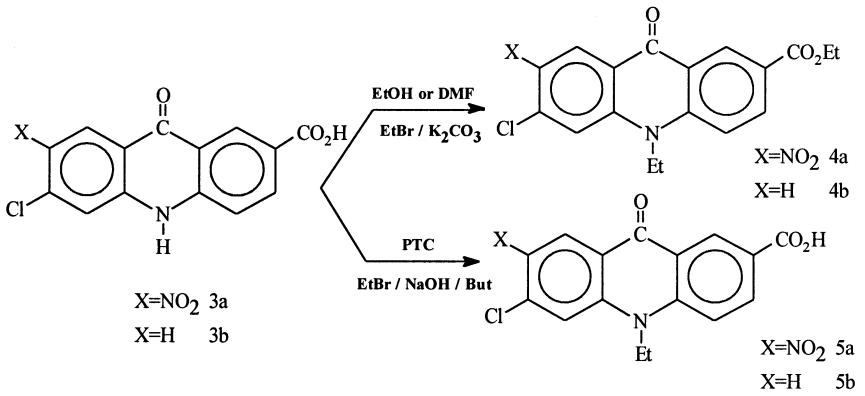
(3c) was oxidised with $KMNO_4$ using solid-liquid phase transfer catalysis in presence of 18-crown-6 as catalyst, yielding the corresponding acridone (3b). The nitration of 2-Carboxy-6-chloro-9(10-H) acridone (3b) using conc. HNO_3 at 40–50°C yielded (3a).

In order to obtain the alkylated compounds (Scheme II), first we used ethanol as protic solvent for the alkylation of 3b with ethyl bromide in presence of potassium carbonate at 50°C during three hours no reaction occurred and only 10% yield was obtained after seven hours reaction time of the corresponding 2-Carboxy-6-chloro-9(10ethyl) acridone ethyl ester (4b).



Scheme II.

To avoid the solvating of a protic solvent, we performed the reaction using DMF as solvent in presence of potassium carbonate during three hours at 50°C to obtain the N-alkylated ethyl ester (4b) as the only reaction product in 67% yield.

When the alkylation reaction was done employing the 7-NO_2 (3a) derivative using condition previously reported in DMF, the corresponding 2-Carboxy-6-chloro-7-nitro-10-(ethyl) acridone ethyl ester (4a) was obtained in 79% yield.

The presence of protic solvents reduces the acridones reactivity because of the formation of intramolecular hydrogen bonding. This does not occurs using DMF as solvent, in this case the compound is free to react with the electrophylic reagent.

The resultant ethyl ester (4a) and (4b) were base hydrolysed using 2N potassium hydroxide solution to give N-ethyl acridone-2-carboxylic acids (5a) and (5b).

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The alkylation reaction of 3a and 3b used liquid-liquid phase transfer catalysis, triethylbenzylammonium chloride (TEBAC) as PTC catalyst, the liquid phase was sodium hydroxide solution (50%) and butanone, the alkyl derivative employed was ethyl bromide and the reaction time 2 hours obtaining the 10-ethyl acridiones 5a and 5b in 86% and 88% respectively as the only reaction product.

EXPERIMENTAL

All solvents were destilled prior to use from an appropriate drying agent. Melting point were determined with a hot stage apparatus and are uncorrected. Elemental analysis were performed within the department. Infrared spectra were recorded as KBr pellets on a Philips PU 9512 spectrometer and expressed in cm^{-1}. Mass spectra were recorded on a Quadrupolar Mass Fisions Instrument TRIO 1000 spectrometer. The ^{1}H and ^{13}C NMR spectra were recorder using DMSO-d_6 as solvent with tetramethylsilane (TMS) as an internal standard on a Bruker A 250 Z NMR spectrometer. The chemical shifts are reported in parts per million (δ ppm). Compound 2b and 3b were reported by Gorving[2] and compound 2c and 3c were reported by Madgison and Grigooruski[18] but no spectral data have been reported. Verification and purity of these compounds were established by combustion analysis and spectral data, using the instrumentation described above.

N-Phenylanthranilic Acid Derivatives, General Procedure

A mixture of benzoic acid (0.02 mol), amine (0.04 mol), potassium carbonate (0.3 mol), copper powder (0.03 g) and DMF (60 mL) was heated and stirred at reflux temperature for 2.5 hours. The reaction mixture was added slowly to a 1:1 (H_2O:HCl) solution (240 mL), the solid obtained was filtered, washed with water and dissolved in a 1% sodium hydroxide solution (260 mL). The acid was precipitated with a (1:3) diluted acetic acid (16 mL), filtered and crystallised to give the corresponding N-phenylanthranilic acid.

2[(4-Carboxyphenyl) amino] 5-chloro-4-nitro benzoic acid (2a) was recrystallyzed from acetic acid/water (1:3) to yield the pure acid as a yellow solid (**3g**; **45%**), **mp** 288–290°C; **IR(cm^{-1})**: 3455.6(NH); 3087.2(OH acid); 1685.7(C = O); 1533.5(as NO_2); 1324.0(sy NO_2) and 767.2(C-Cl); **NMR-^{1}H** δ: 6.95 (2H, dd, $C_{6'}$-H and $C_{2'}$-H); 7.40 (1H, m, C_6-H), 8.0 (2H, d, $C_{3'}$-H and $C_{5'}$-H); 8.65 (1H, s, C_3-H), 10.5 (s, carboxylic acids); **NMR-^{13}C** δ: 166.47 (C acid); 167.55 (C acid) ; 149.35 (C_1); 142.19 (C_1); 136.02 (C_5);

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132.29 (C_4); 130.94 (C_6); 126.99 (C_3); 121.92 ($C_{3'}$y $C_{5'}$); 119.11 ($C_{4'}$); 115.62 (C_2); 112.01 ($C_{6'}$ and $C_{2'}$); **ME (m/z)**: 336 (M^+). **Analysis**: $C_{14}H_9ClN_2O_6$ requires: C, 50.00; H, 2.68; N, 8.33; Cl, 10.56. Found: C, 49.85; H, 2.61; N, 8.26; Cl, 10.47.

2[(4-Carboxyphenyl) amino] 5-chloro benzoic acid (2b) was recrystallyzed from ethanol/water as a yellow product (**4.08 g**; **70%**), **mp**: 284.0–284.2°C. **IR** (cm^{-1}): 3400.5(NH); 3033(OH acid); 1677.8(C = O); 766.6(C-Cl), **NMR-^{1}H** δ: 7.94 (2H, m, $C_{6'}$-H and $C_{2'}$-H); 7.35 (2H, m, C_4-H and C_6-H); 6.73 (2H, dd, $C_{3'}$-H, $C_{5'}$-H); 10.0 (2H, s, carboxylic acids). **NMR-^{13}C** δ: 166.66 (C acid); 168.81 (C acid); 146.02 (C_1); 144.26 ($C_{1'}$); 138.62 (C_5); 133.52 (C_3); 130.99 ($C_{3'}$ and $C_{5'}$); 124.55 (C_6), 119.12 ($C_{4'}$); 118,80 (C_4); 114.37 (C_2); 113.29 ($C_{6'}$ and $C_{2'}$). **ME (m/z)**: 291(M^+). **Analysis**: $C_{14}H_{10}ClNO_4$ requires: C, 57.70; H, 3.44; N, 4.81; Cl, 12.20. Found: C, 57.35; H, 3.46; N, 4.70; Cl, 12.16.

2[(4-Methylphenyl) amino] 5-chloro benzoic acid (2c) was recrystallyzed from acetic acid/water (1:3) to yield the pure acid as a yellow solid (**2.6 g**; **50%**), **mp**: 226–228°C. **IR** (cm^{-1}): 3336(NH); 3033(OH acid); 1677,8(C = O); 748(C-Cl). **NMR-^{1}H** δ: 2.3 (3H, s, CH_3), 6.74 (2H, dd, $C_{6'}$-H and $C_{2'}$-H), 6.95 (2H, d, C_4-H and C_6-H), 7.2 (2H, m, $C_{3'}$-H and $C_{5'}$-H), 7.88 (1H, d, C_3-H) and 13.2 (s, carboxylic acids). **NMR-^{13}C** δ: 169.24 (C acid); 149.05 (C_1); 138.86 (C_4,); 136.63 ($C_{1'}$); 133.75 (C_6); 133.58 ($C_{3'}$ y $C_{5'}$); 130.09 (C_5); 123.25 (C_3); 116.44 (C_4); 111.92 (C_2); 110.43 ($C_{6'}$ and $C_{2'}$) y 39.45 (C methyl). **EM (m/z)**: 261(M^+). **Analysis**: $C_{14}H_{12}ClNO_2$ requires: C, 64.20; H, 4.58; N, 5.35; Cl, 13.57. Found: C, 64.17; H, 4.50; N, 5.38; Cl, 13.41.

Acridones, General Procedure

A Mixture of the appropriate N-phenylanthranilic acid derivatives obtained (2g) was heated in conc. sulphuric acid (10 mL) at 100°C for one hour to produce a fluorescent solution which was diluted with 100 mL of ice-cold water. Solid product that separated was isolated by Buchner filtration and suspended in boiling water (100 mL), filtered hot and dried in a vacuum oven at 100°C for one hour and purified by recrystallization.

2-Carboxy-6-chloro-7-nitro-9(10H) acridone (3a) was obtained as a green solid (**1.9 g; 94.6%**), **mp**: > 350°C. **IR** (cm^{-1}): 3672(NH), 3080 (OH acid), 1728 (C = O), 728 (C-Cl), 1532 (as NO_2), 1332 (sy NO_2). **NMR-^{1}H** δ: 8.8 (1H, d, C_8-H), 8.2 (1H, m C_1-H), 7.6 (1H, d, C_3-H), 7.4 (1H, s, C_5-H), 7.3 (1H, dd, C_4-H), **NMR-^{13}C** δ: 166.38 (C acid); 143.25 (C_{5a}); 141.34 (C_{4a}); 138.27 (C_2); 133.38 (C_3); 132.53 (C_8); 128.31 (C_6); 128.14 (C_{8a}); 123.44 (C_{1a}); 121.95 (C_1); 119.72 (C_7); 117.48 (C_5); 116.42 (C_4). **ME (m/z)**: 318 (M^+).

Analysis: $C_{14}H_7ClN_2O_5$: requries: C, 52.83; H, 2.20; N, 1.80; Cl, 11.16. Found: C, 52.78; H, 1.98; N, 1.83; Cl, 11.24.

2-Carboxy-6-chloro-9(10H) acridone (3b) was obtained as a grey solid **(1.79 g; 95.6%)**, **mp**: > 350°C. **IR (cm^{-1})**: 3628(NH), 3184(OH acid), 1776 (C = O), 708 (C-Cl). **NMR-^{1}H** δ: 8.1 (1H, m, C_1-H); 7.8 (s, 1H, C_8-H), 7.6 (d, 1H, C_3-H), 7.5 (d, 1H, C_7-H), 6.8 (d, 1H, C_5-H) and 6.7 (dd, 1H, C_4-H). **NMR-^{13}C** δ: 167.80 (C acid); 150.0 (C_{5a}); 146.2 (C_{4a}); 139.4 (C_3); 134.7 (C_6); 132.0 (C_1); 131.6 (C_8); 127.1 (C_{8a}); 125.3 (C_{1a}); 119.8 (C_7); 118.0 (C_4); 117.5 (C_5) y 108.1 (C_2). **ME** (**m/z**): 273 (M^+) **Analysis**: $C_{14}H_8ClNO_3$: requires: C, 61.64; H, 2.93; N, 5.10; Cl, 13.00. Found: C, 61.60; H, 2.98; N, 4.89; Cl, 12.90.

2-Methyl-6-chloro-9(10H) acridone (3c) was obtained as a yellow solid (**1.76 g; 95.0%**), **mp**: > 350°C. **IR** (**cm^{-1}**): 3264(NH),1652(C = O), 2920(as CH_3), 2820(sy CH_3). **NMR-^{1}H** δ: 8.2 (1H, d, C_1-H), 8.0 (1H, s, C_8-H), 7.4 (1H, m, C_7-H), 7.3 (1H, d, C_3-H), 7.2 (1H, d, C_4-H), 7.0 (1H, s, C_5-H) y 2.4 (3H, s, CH_3). **NMR-^{13}C** δ: 171.20 (C = O); 138.71 (C_{5a}); 134.92 (C_{4a}); 137.56 (C_6); 130.50 (C_1); 128.10 (C_8); 123.70 (C_7); 124.85 (C_{8a}); 120.78 (C_2); 118.80 (C_{1a}); 117.11 (C_4); 115.95 (C_5) and 20.26 (C methyl). **ME** (**m/z**): 243 (M^+). **Analysis** $C_{14}H_{10}ClNO$: requires C, 68.90; H, 4.10; N, 5.75; Cl, 14.58. Found: C, 68.71; H, 4.09; N, 5.75; Cl, 14.52.

2-Carboxy-6-chloro-9(10H) Acridone (3b) Obtained Oxidising (3c)

A mixture of potassium permanganate 3.16 g (0.02 mol), 6-chloro-2methyl-9 (10H) acridone (3c, 2.43 g, 0.01 mol), benzene (20 mL) and de 18-crown-6 (0.073 g) was stirred and heated to reflux for three hours and then cooled to room temperature. A 1% sodium hydroxide solution (20 mL) was added and the mixture heated and stirred for one hour. The aqueous layer is acidified using diluted HCl:H_2O (1:1) and the precipitated product filtered and identified as 2-carboxy-6-chloro-9(10-H) acridone (3b) (2.05; 75%).

2-Carboxy-6-chloro-7-nitro-9(10H) Acridone. Nitration of (3b)

A mixture of 2-carboxy-6-chloro-9(10H) acridone (3b, 2.0 g) and 70% nitric acid (10 mL) was heated in a water bath to 40–50°C stirred for two hours at room temperature. The reaction mixture was then poured into water (120 mL), the resulting precipitated was recovered in a 5% Na_2CO_3 solution. Concentration of the solution gave a solid which was filtered and

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redisolved in water (70 mL). The solution was acidified and dried to yield 40% of 2-carboxy-6-chloro-7-NO_2-9 (10-H) acridone.

Acridone Alkylation (using solvent)

A mixture of acridone (0.01 mol), potassium carbonate (0.03 mol), ethyl bromide (0.8 mL) in appropriated solvent (20 mL) was stirred with heating for three hours at 50°C. The reaction mixture was added slowly to a diluted HCl solution, the solid obtained was filtered and washed with water, crystallised from ethanol and dried in a vacuum oven at 100°C.

Acridone Alkylation (using PTC)

A mixture of acridone (0.01 mol), 50% sodium hydroxide solution, triethylbenzylammonium chloride 0.08 g, ethyl bromide 0.8 mL and butanone 20 mL was heated and stirred to reflux two hours. The organic layer was separated and dried with anhydrous sodium sulphate. The organic layer is rotoevaporated, the solid obtained is dissolved in water and precipitated with a 1:1 hydrochloric acid:water solution obtaining the corresponding 2-carboxy-N-ethyl acridones.

2-Carboxy-6-chloro-9(10ethyl) acridone ethyl ester (4b) was obtained as a beige solid (**2.20 g; 67,0%), mp**: 202–204°C. **IR** (**cm^{-1}**): 1460(C-H), 1712(C = O), 1608, 1500(C = C), 1225-950(C-H), 828(C-H), 728(C-Cl). **ME** (**m/z**): 329 (M^+). **NMR-^{1}H** δ: 8.2 (1H, d, C_1-H); 8.3 (1H, d, C_3-H); 7.8 (1H, s, C_8-H); 6.9 (1H, d, C_4-H); 6.7 (1H, s, C_5-H). **NMR-^{13}C** δ: 149.8 (C_{5a}); 146.25 (C_{4a}); 135.4 (C_6); 134.8 (C_3); 132.4 (C_8); 130.8 (C_1); 129.5 (C_7); 127.62 (C_{8a}); 125.3 (C_{1a}); 120.3 (C_2); 118.0 (C_5) y 117.8 (C_4). **Analysis:** $C_{18}H_{16}NO_3Cl$: requires: C, 65.65; H, 4.86; N, 4.25; Cl, 10.79. Found: C, 65.40; H, 4.29; N, 4.74; Cl, 10.60.

2-Carboxy-6-chloro-7-nitro-9(10ethyl) acridone ethyl ester (4a) was obtained as a brown solid (**2.95 g; 79%**), **mp**: 199–200°C, **IR** (**cm^{-1}**): 1734(C = O), 1652(C = O), 1596(as NO_2), 1352(sy NO_2), 2920 and 2818 (CH_3). **NMR-^{1}H** δ: 8.45 (C_8-H); 8.23 (C_1-H); 8.0 (C_3-H); 7,0 (C_5-H); 6.65 (C_4-H). **NMR-^{13}C** δ: 152.0 (C_7); 149.2 (C_{5a}); 149.14 (C_{4a}); 134.0 (C_3); 132.1 (C_1); 130.2 (C_6); 127.9 (C_{8a}); 127.5 (C_8); 126.2 (C_{1a}); 120.5 (C_2); 119.0 (C_5) y 117.2 (C_4). **ME** (**m/z**): 374(M^+). **Analysis**: $C_{18}H_{15}N_2O_5Cl$: requires C, 57.67; H, 4.01; N, 7.49; Cl, 9.49. Found: C, 54.41; H, 4.12; N, 7.52; Cl, 9.54.

2-Carboxy-6-chloro-9(10ethyl) acridone (5b) was obtained as a cream solid (**2.63 g; 87.5%**), **mp:** 201–203°C. **IR** (**cm^{-1}**): 2936 (OH acid), 1460 (C = C), 1732(C = O), 1668(C = O), 728(C-Cl). **NMR-^{1}H** δ: 11.0 (1H, d, H

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acid); 8.3 (1H, d, C_1-H); 8.0 (1H, s, C_3-H); 7.5 (1H, s, C_5-H); 7.3 (1H, d, C_5-H); 7.0 (1H, d, C_7-H); 6.7 (1H, s, C_5-H). **NMR-^{13}C** δ: 150.0 (C_{5a}); 146.0 (C_{4a}); 135.32 (C_6); 134.65 (C_3); 132.62 (C_1); 132.3 (C_8); 129.7 (C_7); 127.9 (C_{8a}); 125.1 (C_{1a}); 120.1 (C_2); 118.2 (C_5) and 117.4 (C_4). **ME** (**m/z**): 301 (M^+) **Analysis**: $C_{16}H_{12}NO_3Cl$: C, 63.79; H, 3.99; N, 4.65; Cl, 11.79. Found: C, 63.41; H, 4.10; N, 4.74; Cl, 12.04.

2-Carboxy-6-chloro-7-nitro-9-(10ethyl) acridone (5a) was obtained as a green solid (**2.79 g**; **85.9%**), **mp**: $>350^{\circ}$C. **IR** (**cm^{-1}**): 2928 (OH acid), 1460(C = C), 1688(C = O), 1652 (C = O), 1546 and 1352 NO_2, 728(C-Cl). **NMN-^{1}H** δ: 12.0 (H acid); 8.5 (1H, d, C_1-H); 8.3 (1H, d, C_3-H); 8.0 (1H, d, C_7-H); 7.1 (1H, s, C_5-H); 7.0 (1H, d, C_4-H). **NMR-^{13}C** δ: 152.2 (C_{5a}); 151.5 (C_{4a}); 148.0 (C_7); 134.7 (C_3); 132.50 (C_1); 130.6 (C_6); 127.8 (C_{8a}); 127.1 (C_8); 126.0 (C_{1a}); 119.7 (C_2); 119.3 (C_5) y 117.4 (C_4). **ME** (**m/z**): 346 (M^+). **Analysis**: $C_{15}H_{11}N_2O_5Cl$: C, 52.02; H, 3.18; N, 8.09; Cl, 10.26. Found: C, 52.21; H, 3.30; N, 8.24; Cl, 10.30.

CONCLUSIONS

In the synthesis of N-alkyl derivatives of acridones 2-carboxylic acids the best results were obtained using PTC conditions.

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