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Reactions of some nitrogen heterocycles with chlorodifluoromethane under conditions of phase-transfer catalysis

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Abstract

Aromatic heterocyclic compounds containing one to four nitrogen atoms react with chlorodifluoromethane in the presence of concentrated aqueous sodium hydroxide and a catalyst, benzyltriethylammonium chloride (TEBAC) in dichloromethane or THF (phase-transfer catalysis, PTC) with formation of N-difluoromethyl substituted derivatives. The process takes place by reaction of N-anions from these heterocycles with difluorocarbene and protonation of difluoromethyl anions thus formed.

Keywords: Phase-transfer catalysis; Difluorocarbene; N-difluoromethylation; Nitrogen heterocycles

1. Introduction

Chlorodifluoromethane (**1**) treated with a base generates difluorocarbene [1,2], possibly without intervention of chlorodifluoromethyl anion [3]. This electrophilic carbene generated from **1** under different basic conditions has been utilized for difluoromethylation of nitrogen heterocycles. Thus, the reaction with pyrazole, benzopyrazoles or perimidines was carried out in aqueous-acetone solution of potassium hydroxide [4], with 4-ethoxycarbonylpyrazole in the presence of solid potassium carbonate in DMF [5], with phenylazoles with sodium hydride in THF or DMF [6], and with 2-mercaptoazoles with potassium hydroxide in *i*-propanol–water mixture or DMF [7]. In the case of indoles, the process was performed with sodium *t*-butoxide [8] or sodium hydride in DMF [9]; the former base was also applied for the reaction of carbostyril, giving mixture of N- and O-derivatives [8]. Similarly, quinoxalin-2-ones afforded mixtures of N- and O-difluoromethylated compounds in moderate yield when solid potassium carbonate or casium fluoride in DMF were applied [10], while the reaction of spiro[imidazolidine(quinazoline)]trione with sodium hydride in DMF led to formation only of the N-derivative [11].

Little is known about the application of alkali metal hydroxides and a quaternary ammonium salt as a catalyst (phase-transfer catalysis, PTC [12–14]) for difluoromethylation of nitrogen heterocycles with **1**. The preparation of N-difluoromethylated herbicides from aryltriazolinones, under these conditions, was patented [15,16].

2. Results and discussion

Chlorodifluoromethane (**1**) is not suitable for the preparation of *gem*-difluorocyclopropanes from alkenes under PTC conditions [17,18] since difluorocarbene generated from **1** by a concerted α-elimination [3] at the interface of the two-phase system, hydrolyzed at a high rate [19]. However, literature data indicate that even large amounts of water in the basic system do not disturb the reaction of **1** with nitrogen heterocycles, and N-difluoromethylated products were formed in satisfactory yields [4,7].

These results encouraged us to investigate reaction of aromatic heterocycles which posses one to four nitrogen atoms **2a–k** and **6** with **1**, carried out in the presence of 50% aqueous sodium hydroxide and benzyltriethylammonium chloride (TEBAC) as a catalyst in dichloromethane or THF (liquid–liquid variant of PTC).

Reaction of indole (**2a**) with **1** led to formation of N-derivative **3a** (yield 50%), while the products from

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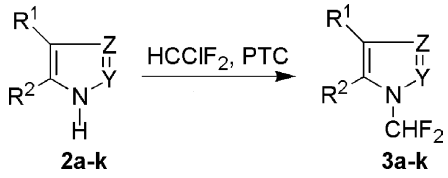
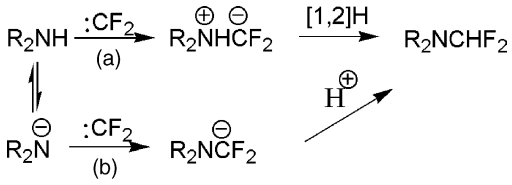


Scheme 1.



Scheme 3.

3-methylindole and carbazole were insufficiently stable for isolation either by column chromatography or by vacuum distillation. According to the literature, when 2,3-dimethylindole was allowed to react with ethyl chlorodifluoroacetate and potassium *t*-butoxide, 3-difluoromethyl-2,3-dimethyl-3H-indole was formed [20]. Under the conditions studied, pyrrole formed by PTC the product in ca. 8% yield (determined by GC).

Series of aromatic diazaheterocycles **2b–j** reacted with chlorodifluoromethane (**1**) affording N-difluoromethyl substituted derivatives **3b–j**, often in good yields. In the case of derivatives possessing, a hydroxy group **2i,j** O-difluoromethylation competed with N-difluoromethylation, giving the products **3i,j** and disubstituted derivatives **4** and **5**. Bis-difluoromethylation was already noticed during reactions of 2-mercaptoazoles [6]. Amide **2f** was exclusively difluoromethylated at the ring nitrogen atom. Acetyl derivative **2g** was rather unstable under PTC conditions; hence, the yield of **3g** was only 13%.

Chlorodifluoromethane (**1**) reacted smoothly with benzotriazole (**2k**) and less easily with 5-benzyl-1H-tetrazole (**6**) giving the products **3k** or **7** in 72 and 38% yield, respectively (Schemes 1 and 2). In the latter case, a mixture of regioisomers resulted, to which structures **7a** and **7b** have been tentatively ascribed (NOE experiments did not solve this problem).

All reactions of **1** with nitrogen heterocycles were carried out under mild conditions, at 20–40 °C, and occurred at a rather high rate.

Two mechanistic pathways leading to N-difluoromethylation of heterocycles have been considered [4,6,21]: (a) reaction of difluorocarbene with p-electron pair of nitrogen atom leading to a N-ylide which further enters [1,2]H shift or (b) its reaction with a N-anion affording the basic N-difluoromethyl anion which gave the product after protonation (Scheme 3).

It is well established that the PTC conditions are sufficiently basic to cause deprotonation of a variety of azoles [22,23]; hence, mechanistic pathway (b) seems more plausible. When a weak N–H acidic compound, not able to generate a N-anion under PTC conditions is allowed to react with **1**, N-formylation occurred, e.g. morpholine gave N-formyl derivative in good yield. In this case mechanistic pathway (a) is involved and hydrolysis of N-difluoromethyl into N-formyl derivative is facilitated by the presence of highly basic nitrogen atom. To confirm the pathway (b), reaction of **2h** with **1** was carried out under PTC conditions, in excess of carbon tetrachloride. This reagent chlorinates carbanions easily by an halophilic process [24,25]. The reaction studied afforded 1-chlorodifluoromethyl-2-phenylbenzimidazole (**8**) in 21% yield apart from the product **3h** (Scheme 4).

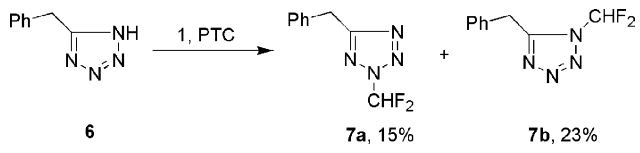
This product is not formed when **3h** was allowed to react with carbon tetrachloride under PTC conditions. The results described above testify that halogenation of N-difluoromethyl anion competed with its protonation.

A liquid–liquid variant of PTC is a convenient tool for N-difluoromethylation of a variety of aromatic nitrogen heterocycles with chlorodifluoromethane (**1**) via reaction of their N-anions with difluorocarbene. The procedure is simple and the process is rather fast affording the products often in good yields.

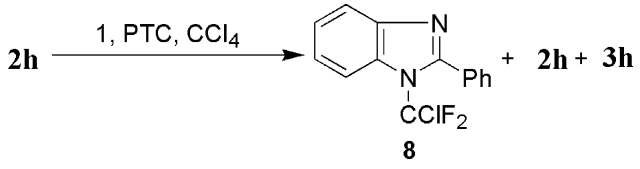
3. Experimental

3.1. General

Melting points were measured on a capillary melting point apparatus and are uncorrected. ^{1}H NMR spectra were recorded on a Varian Mercury (at 400 MHz) or Varian Gemini (at 200 MHz), ^{13}C NMR spectra on a Varian Mercury (at 100 MHz) spectrometers in $CDCl_3$ with tetramethylsilane as internal reference. ^{19}F NMR spectra were recorded on a Varian Mercury (at 376 MHz) in $CDCl_3$ with trifluoroacetic acid as internal reference. All chemical shifts are reported in ppm ($\delta = 0.00$). The abbreviations used are as follows: s, singlet; d, doublet; t, triplet; q, quartet; m, multiplet. Gas chromatography (GC) analyses were carried out on a gas chromatograph, Agilent 6850 Series GC System equipped with HP-50+ (30 m) column. Microanalyses were obtained using a CHN/S Perkin-Elmer 2400 element analyzer. Column chromatography was performed using Merck basic aluminiumoxid 90 (70–230 mesh) with hexane and ethyl acetate mixtures (gradient) as eluents. Chlorodifluoromethane



Scheme 2.



Scheme 4.

(**1**) and heterocycles **2a–d**,**h** were commercial materials, while benzimidazoles **2e** [26], **2f** [27], **2g** [28], **2i**,**2j** [29], benzotriazole **2k** [30] and benzyltetrazole **6** [31] were prepared by literature procedures.

3.2. General procedure for N-difluoromethylation of nitrogen heterocycles (*2a–d*)

Into a three-necked, round-bottomed flask equipped with reflux condenser, mechanical stirrer and glass pipe for introducing **1**, heterocycle **2a–d** (40 mmol), 50% aq. NaOH (6.4 ml, 9.6 g, 120 mmol), TEBAC (0.454 g, 2 mmol) and CH_2Cl_2 or THF (ca. 50 ml) were placed. The content of the flask was stirred for ca. 2 min, then chlorodifluoromethane (**1**) was bubbled through the mixture for 0.5–3 h, and the progress of the reaction was monitored by GC. The mixture was diluted with CH_2Cl_2 (50 ml), the organic phase was decanted from semisolid inorganic one which sticks to the wall of the reaction flask, and dried over $MgSO_4$. The solvent was evaporated under reduced pressure and the residue was purified by vacuum distillation (Table 1).

3.3. General procedure for N-difluoromethylation of nitrogen heterocycles (*2e–k*) and (*6*)

The reactions were carried out as described in p. **3.1** starting from heterocycle **2e–k** or **6** (6 mmol), 50% aq. NaOH (0.96 ml, 1.44 g, 18 mmol), TEBAC (68 g, 0.3 mmol) and THF (20 ml). The mixture was diluted with CH_2Cl_2 (20 ml), the organic phase was decanted, worked up and the product was isolated by column chromatography (Table 1).

3a, liquid, b.p. 76–78 °C/3.5 Torr, ^{1}H NMR δ 6.65 (d, J = 3.2 Hz,1H), 7.27 (t, J = 61 Hz, 1H, C**H**F_2), 7.29 (d, J = 3.6 Hz, 1H),, 7.24–7.31 (m, 2H), 7.56–7.58 (m, 1H), 7.64–7.66 (m, 1H). ^{13}C NMR δ 105.9, 110.0 (t, J = 246 Hz, **C**HF_2), 110.6, 121.4, 121.9, 123.1, 129.6, 134.0. ^{19}F NMR δ –95.7 (d, J_{H-F} = 61 Hz, 2F). HRMS calcd. for $C_9H_7NF_2$: 167.0546. Found: 167.0544.

3b, liquid, b.p. 62–63 °C/15 Torr, literature [3] b.p. 155–156 °C/740 Torr, ^{1}H NMR δ 7.10 (t, J = 61 Hz, 1H, C**H**F_2), 7.15 (s, 1H), 7.20 (t, J = 1.4 Hz, 1H), 7.81 (s, 1H). ^{13}C NMR δ 108 (t, J = 251 Hz, **C**HF_2), 114.9, 130.9, 134.4. ^{19}F NMR δ −96.2 (d, J_{H-F} = 61 Hz, 2F). HRMS calcd. for $C_4H_4N_2F_2$: 118.0342. Found: 118.0343.

3c1, liquid, b.p. 65–66 °C/10 Torr, ^{1}H NMR δ 2.21 (d, J = 0.4 Hz, 3H), 6.79 (t, J = 1.2 Hz, 1H), 7.02 (t, J = 61 Hz, 1H, C**H**F_2), 7.67 (d, J = 1.2 Hz, 1H). ^{13}C NMR δ 8.6, 108.0 (t, J = 248 Hz, **C**HF_2),110.7, 128.1, 133.7. ^{19}F NMR δ −96.6 (d, J_{H-F} = 61 Hz, 2F).

3c2, liquid, b.p. 65–66 °C/10 Torr, ^{1}H NMR δ 2.30 (d, J = 0.8 Hz, 3H), 6.89 (s, 1H), 7.00 (t, J = 60 Hz, 1H, C**H**F_2), 7.70 (s, 1H). ^{13}C NMR δ 12.7, 108.5 (t, J = 248 Hz, **C**HF_2), 110.7, 128.1, 134.6. ^{19}F NMR δ −98.3 (d, J_{H-F} = 60 Hz, 2F). HRMS calcd. for $C_5H_6N_2F_2$: 132.0499. Found: 132.0495.

3d, solid, b.p. 67–68 °C/0.3 Torr, literature [3] b.p. 102–103 °C/2 Torr, m.p. 40–41 °C, literature [3] m.p. 42–43 °C, ^{1}H NMR δ 7.37 (t, J = 60 Hz, 1H, C**H**F_2) 7.35–7.40 (m, 2H), 7.59–7.62 (m, 1H), 7.82–7.84 (m, 1H), 8.16 (s, 1H). ^{13}C NMR δ

Table 1
Difluoromethylation of nitrogen heterocycles **2a–k** with chlorodifluoromethane (**1**) under PTC conditions

Nos. 2 and 3	R^1	R^2	Y	Z	Temperature (°C)	Solvent	Product	
							Purity[a] (%)	Yield (%)
a	CH=CH–CH=CH		CH	CH	40	CH_2Cl_2	96	50[b]
b	H	H	CH	N	20	CH_2Cl_2	99	40[b]
c	Me	H	CH	N	40	CH_2Cl_2	99	46[b,c]
d	CH=CH–CH=CH		CH	N	20	THF	97	65[b]
e	CH=CH–CH=CH		CMe	N	20	THF	99	67[d]
f	CH=CH–CH=CH		$CCONH_2$	N	20	THF	98	69[d]
g	CH=CH–CH=CH		CCOMe	N	20	THF	96	13[d]
h	CH=CH–CH=CH		CPh	N	20	THF	ca. 100	91[d]
i	CH=CH–CH=CH		CCH(OH)Me	N	20	THF	95	50[d,e]
j	CH=CH–CH=CH		CCH(OH)Ph	N	20	THF	98	36[d,f]
k	CH=CH–CH=CH		N	N	20	THF	99	72[d]

[a] Determined by GC.
[b] Isolated by vacuum distillation.
[c] Mixture of 1-difluoromethyl-4-methylimidazole (**3c1**) and 1-difluoromethyl-5-methylimidazole (**3c2**), ratio 2:1.
[d] Isolated by column chromatography.
[e] Additionally, 1-difluoromethyl-2-(1-difluoromethoxy)ethylbenzimidazole (**4**) was isolated in 11% yield (purity 99%).
[f] Additionally, 1-difluoromethyl-2-(1-difluoromethoxy)benzylbenzimidazole (**5**) was isolated in 28% yield (purity 96%).

108.9 (t, J = 248.4 Hz, 1C, **C**HF_2), 111.1, 120.6, 124.2, 124.8, 130.3, 139.1, 143.4. ^{19}F NMR δ −98.6 (d, J_{H-F} = 60 Hz, 2F). Anal. calcd. for $C_8H_6F_2N_2$: C, 57.2, H, 3.6, N, 16.7. Found: C, 57.3, H, 3.5, N, 16.7.

3e, solid, m.p. 61–62 °C, literature [3] m.p. 62–63 °C, ^{1}H NMR δ 2.70 (s, 3H), 7.28 (t, J = 59 Hz, 1H, C**H**F_2), 7.27–7.33 (m, 2H), 7.51–7.55 (m, 1H), 7.68–7.72 (m, 1H). ^{13}C NMR δ 14.5, 108.8 (t, J = 246.5 Hz, 1C, **C**HF_2), 110.5, 119.7, 123.7, 123.8, 132.0, 142.5, 149.4. ^{19}F NMR δ –99.2 (d, J_{H-F} = 58 Hz, 2F). Anal. calcd. for $C_9H_8F_2N_2$: C, 59.3, H, 4.4, N, 15.4. Found: C, 59.3, H, 4.5, N, 15.4.

3f, solid, m.p. 182–184 °C, ^{1}H NMR δ 6.03 (s, 1H), 7.42–7.51 (m, 2H), 7.63 (s, 1H), 7.80–7.85 (m, 2H), 8.77 (t, J = 59 Hz, 1H, C**H**F_2). ^{13}C NMR δ 109.5 (t, J = 249.5 Hz, 1H, **C**HF_2), 113.8, 121.3, 125.0, 126.7, 132.6, 141.1, 160.6. ^{19}F NMR δ −100.6 (d, J_{H-F} = 59 Hz, 2F). Anal. calcd. for $C_9H_7F_2N_3O$: C, 51.2, H, 3.3, N, 19.9. Found: C, 51.2, H, 3.4, N, 19.8.

3g, liquid, ^{1}H NMR δ 2.86 (s, 3H), 7.43–7.53 (m, 2H), 7.80–7.83 (m, 1H), 7.90–7.93 (m, 1H), 8.49 (t, J = 59 Hz, 1H, C**H**F_2). ^{13}C NMR δ 27.8, 109.4 (t, J = 249.6 Hz, 1C, **C**HF_2), 113.9, 122.3, 125.2, 127.8, 130.9, 141.6, 144.4, 192.9. ^{19}F NMR δ −100.0 (d, J_{H-F} = 59 Hz, 2F). Anal. calcd. for $C_{10}H_8F_2N_2O$: C, 57.2, H, 3.8, N, 13.3. Found: C, 57.3, H, 3.9, N, 13.5.

3h, solid, m.p. 35–37 °C, ^{1}H NMR δ 7.29 (t, J = 58 Hz, 1H, C**H**F_2), 7.39–7.42 (m, 2H), 7.56–7.589 (m, 3H), 7.72–7.87 (m, 4H). ^{13}C NMR δ 110.1 (t, J = 246.8 Hz, 1C, **C**HF_2), 112.7, 120.4, 124.3, 124.5, 128.6, 129.2, 129.4, 130.9, 131.7, 142.9, 151.8. ^{19}F NMR δ −99.1 (d, J_{H-F} = 58 Hz, 2F). Anal. calcd. for $C_{14}H_{10}F_2N_2$: C, 68.9, H, 4.1, N, 11.5. Found: C, 68.8, H, 4.1, N, 11.5.

3i, solid, m.p. 65–67 °C, ^{1}H NMR δ 1.51 (m,3H), 5.07 (q, J = 6.9 Hz, 1H), 5.99 (s, 1H, O**H**), 7.15–7.64 (m, 4H), 8.02 (t, J = 59 Hz, NC**H**F_2). ^{13}C NMR δ 22.1, 64.6, 109.3 (t, J = 246.5 Hz, 1C, N**C**HF_2), 112.6, 119.4, 123.9, 124.6, 131.6, 141.1, 154.9. ^{19}F NMR δ −100.6, –101.5 (part AB of ABX, J_{F-F} = 229.3 Hz, J_{H-F} = 59 Hz, 2F, NCH**F**$_2$). Anal. calcd. for $C_{10}H_{10}F_2N_2O$: C, 56.6, H, 4.8, N, 13.2. Found: C, 56.5, H, 4.7, N, 13.1.

4, liquid, ^{1}H NMR δ 1.82 (d, J = 1.7 Hz, 3H), 5.71 (q, J = 6.8 Hz, 1H), 6.35, (t, J = 73 Hz, 1H, –OC**H**F_2), 7.36–7.39 (m, 2H), 7.68 (t, J = 59 Hz, 1H, –NC**H**F_2), 7.69–7.79 (m, 2H). ^{13}C NMR δ 20.9, 67.9, 108.9 (t, J = 246.9 Hz, 1C, N**C**HF_2), 112.6, 115.7 (t, J = 261 Hz, 1C, O**C**HF_2), 120.6, 124.2, 125.1, 132.0, 141.9, 150.1. ^{19}F NMR δ −87.6, −88.1 (part AB of ABX, J_{F-F} = 161.7 Hz, J_{H-F} = 73 Hz, 2F, OCH**F**$_2$), −99.3, −100.6 (part AB of ABX, J_{F-F} = 228.6 Hz, J_{H-F} = 59 Hz, 2F, NCH**F**$_2$). Anal. calcd. for $C_{11}H_{10}F_4N_2O$: C, 50.4, H, 3.8, N, 10.7. Found: C, 50.3, H, 3.8, N, 10.7.

3j, solid, m.p. 126–128 °C, ^{1}H NMR δ 6.27 (s, 1H), 6.69 (s, 1H), 7.25–7.36 (m, 7H), 7.57–7.61 (m, 2H), 7.63 (t, J = 58 Hz, 1H, –NC**H**F_2). ^{13}C NMR δ 69.9, 108.9 (t, J = 247.2 Hz, 1C, N**C**HF_2), 112.7, 119.7, 124.1, 124.8, 125.4, 128.3, 128.8, 131.7, 138.7, 141.1, 153.7. ^{19}F NMR δ −91.9, −92.4 (part AB of ABX, J_{F-F} = 213.4 Hz, J_{H-F} = 58 Hz, 2F, NCH**F**$_2$). Anal. calcd. for $C_{15}H_{12}F_2N_2O$: C, 65.7, H, 4.4, N, 10.2. Found: C, 65.9, H, 4.8, N, 10.0.

5, liquid, ^{1}H NMR δ 6.52 (t, J = 73 Hz, 1H, OC**H**F_2), 6.73 (s, 1H), 7.35 (t, J = 58 Hz, 1H, NC**H**F_2), 7.34–7.42 (m, 7H), 7.40–7.85 (m, 2H). ^{13}C NMR δ 72.9, 108.8 (t, J = 247.2 Hz, 1H N**C**HF_2), 112.8, 115.9 (t, J = 262 Hz, 1H, O**C**HF_2), 120.8, 124.3, 125.3, 125.6, 129.1, 129.2, 132.1, 134.8, 142.1, 148.9, 166.6. ^{19}F NMR δ –86.8, –89.4 (part AB of ABX, J_{F-F} = 161.1 Hz, J_{H-F} = 73 Hz, 2F, OCH**F**$_2$), –100.3, −101.6 (part AB of ABX, J_{F-F} = 226.7 Hz, J_{H-F} = 58 Hz, 2F, NCH**F**$_2$). Anal. calcd. for $C_{16}H_{12}F_4N_2O$: C, 59.3, H, 3.7, N, 8.6. Found: C, 59.3, H, 3.7, N, 8.7.

3k, solid, m.p. 38–40 °C, ^{1}H NMR δ 7.46–7.50, (m, 1H), 7.59–7.64 (m, 1H), 7.81 (d, J = 8.4 Hz, 1H), 7.86 (t, J = 58 Hz, 1H, C**H**F_2), 8.11–8.13 (m, 1H). ^{13}C NMR δ 110.7, 111.3 (t, J = 249.5 Hz, 1C, **C**HF_2), 120.4, 125.5, 129.4, 129.9, 146.4. ^{19}F NMR δ −101.9 (d, J_{H-F} = 58 Hz, 2F). Anal. calcd. for $C_7H_5F_2N_3$: C, 49.7, H, 3.0, N, 24.8. Found: C, 49.8, H, 3.1, N, 24.6.

7a, liquid, ^{1}H NMR δ 4.34 (s, 2H), 7.28–7.36 (m, 5H), 7.58 (t, J = 56 Hz, 1H, C**H**F_2). ^{13}C NMR δ 31.7, 108.7 (t, J = 258.2 Hz, 1C, **C**HF_2), 127.2, 128.7, 128.8, 135.4, 167.1. ^{19}F NMR δ −102.6 (d, J_{H-F} = 58 Hz, 2F). Anal. calcd. for $C_9H_8F_2N_4$: C, 51.4, H, 3.8, N, 26.7. Found: C, 51.4, H, 3.8, N, 26.7.

7b, liquid, ^{1}H NMR δ 4.43 (s, 2H), 7.28–7.37 (m, 5H), 7.57 (t, J = 58 Hz, 1H, C**H**F_2). ^{13}C NMR δ 29.6, 108.7 (t, J = 255.9 Hz, 1C, **C**HF_2), 127.8, 128.7, 128.9, 132.9, 153.7. ^{19}F NMR δ −91.2 (d, J_{H-F} = 58 Hz, 2F). Anal. calcd. for $C_9H_8F_2N_4$: C, 51.4, H, 3.8, N, 26.7. Found: C, 51.3, H, 3.9, N, 26.5.

*3.4. Preparation of 1-chlorodifluoromethyl-2-phenylbenzimidazole (**8**)*

To three-necked, round-bottomed flask equipped with reflux condenser, mechanical stirrer and glass pipe for introducing **1**, imidazole **2h** (970 mg, 5 mmol), 50% aq. NaOH (0.8 ml, 1.2 g, 15 mmol), TEBAC (57 mg, 0.25 mmol), CCl_4 (20 ml) and THF (10 ml) were placed. The content of the flask was stirred for ca. 2 min, then **1** was bubbled through the reaction mixture at ca. 20 °C for 3 h and at 50 °C for 2 h. The mixture was diluted with CH_2Cl_2 (20 ml), worked up as described in p. **3.1** (GC indicated 47% of **8**, 27% of **3h** and 24% of unreacted **2h**) and the product was isolated by column chromatography.

8, liquid, ^{1}H NMR δ 7.49–7.53 (m, 5H), 7.66–7.92 (m, 3H), 7.84–7.86 (m, 1H). ^{13}C NMR δ 112.9, 120.6, 121.3 (t, J = 290 Hz, 1C, **C**HF_2), 124.7, 125.1, 128.1, 129.5, 130.2, 130.3, 132.3, 142.7, 151.1. ^{19}F NMR δ −35.6 (s, 2F). Anal. calcd. for $C_{13}H_9ClF_2N_2$: C, 60.3, H, 3.3, N, 10.1, Cl, 12.7. Found: C, 60.3, H, 3.4, N, 10.0, Cl, 12.7.

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