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PERFLUOROALKYLBENZENESULFONYL CHLORIDES, PERFLUOROALKYLNAPHTHALENESULFONYL CHLORIDES, AND THEIR METAL SALTS

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SUMMARY

Meta-perfluoro-n-alkylbenzenesulfonyl chlorides and 1-perfluoro-n-alkylnaphthalene-5-sulfonyl chlorides were synthesized from the corresponding perfluoroalkylaromatics in good yield by the action of chlorosulfonic acid. From the acid chlorides, salts (RSO_3M, where M = Li, K, Ba, Zn and Al) were prepared and their TGA curves obtained. With the exception of the aluminum salts, the materials were stable to at least 350°C. All the salts were essentially insoluble in $Cl(CFClCF_2)_{3-4}Cl$.

INTRODUCTION

Telomers of chlorotrifluoroethylene, $Cl(CFClCF_2)_xCl$ where x = 3-4, are being developed as nonflammable hydraulic fluids for aircraft systems [1]. These fluids require an anti-corrosion additive. For low temperature applications, <135°C, barium dinonylnaphthalene sulfonate is adequate but it undergoes decomposition at higher temperatures. Furthermore, there are solubility problems and the barium salt is susceptible to

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carbonate precipitation. Introduction of a perfluoroalkyl chain onto the aromatic ring would be expected to enhance the solubility and thermal stability of the resultant sulfonic acid salt whereas replacement of barium by another metal should alleviate the solution instability.

Perfluoroalkyl-substituted aromatics are most readily prepared by the reaction of perfluoroiodoalkanes with either iodo- or bromo-aromatics in polar solvents in the presence of copper. The procedure was originally developed by McLoughlin and Thrower [2] and used successfully by others [3-5]. A large number of perfluoroalkyl-substituted phenyl compounds have been reported [4-8], including a series of naphthalenes. Unfortunately, in the case of the latter, in particular compounds having longer perfluoroalkyl chains, only very limited physical and spectral data were given. Also, since n-perfluoroiodoalkanes were exclusively employed (in the current investigation), differences in melting points and other properties would be anticipated. The route chosen for the preparation of the salts was based on utilizing the perfluoroalkylaromatics as starting materials for the sulfonyl chloride synthesis followed by salt formation.

RESULTS AND DISCUSSION

Reaction of iodobenzene and 1-iodonaphthalene with 1-iodoperfluoro-n-heptane and 1-iodoperfluoro-n-octane in the presence of copper bronze using dimethylsulfoxide (DMSO) as solvent, following essentially the procedure of McLoughlin and Thrower [2], resulted in high yields of the perfluoroalkyl aromatics. These data are summarized in Table 1. Attempts were made to utilize 1-bromonaphthalene instead of the significantly more costly 1-iodonaphthalene. The yields attained were greatly reduced. Increase in reaction times failed to result in further reaction. Actually, beyond 34 h at 125°C, no increase in 1-perfluoro-n-heptylnaphthalene was observed. The increased exposure promoted further production of methylthionaphthalene (identified by GC/MS; 174 $[M]^+$, 159 $[M - CH_3]^+$).

TABLE 1

Fluoroalkylation Reactions

Iodofluoroalkane type (mmol)	Haloaromatic type (mmol)	Copper mmol	Solvent type (mL)	Conditions		Products	Yield %
				Time h	Temp °C		
$n\text{-}C_7F_{15}I$ (54.44)	C_6H_5I (48.52)	156	DMSO[a] (100)	23	125	$n\text{-}C_7F_{15}C_6H_5$[b]	78
$n\text{-}C_8F_{17}I$ (71.41)	C_6H_5I (64.70)	204	DMSO (100)	15	130	$n\text{-}C_8F_{17}C_6H_5$	83
$n\text{-}C_7F_{15}I$ (66.84)	$C_{10}H_7I$[c] (59.28)	189	DMSO (100)	48	125	$n\text{-}C_7F_{15}C_{10}H_7$	89
$n\text{-}C_7F_{15}I$ (5.32)	$C_{10}H_7Br$[d] (4.83)	12.6	DMSO (8)	34	125	$n\text{-}C_7F_{15}C_{10}H_7$ $CH_3SC_{10}H_7$[e]	45 ~40
$n\text{-}C_7F_{15}I$ (10.62)	$C_{10}H_7Br$[d] (9.66)	15	C_5H_5N (15)	72	110	$n\text{-}C_7F_{15}C_{10}H_7$ $C_{10}H_8$[e]	trace ~20
$n\text{-}C_8F_{17}I$ (61.19)	$C_{10}H_7I$[c] (55.58)	189	DMSO (100)	16	120	$n\text{-}C_8F_{17}C_{10}H_7$	96

a Dimethylsulfoxide.

b bp 89-91°C/17 mm Hg; Ref. [2] *bp* 88-90°C/16 mm Hg.

c 1-Iodonaphthalene.

d 1-Bromonaphthalene.

e Identified by mass spectroscopy.

Utilization of pyridine as solvent, based on the literature claims regarding its advantages in naphthalene systems [4], gave only a trace of 1-perfluoro-n-heptylnaphthalene; the major product was naphthalene.

The synthesis of aromatic sulfonic acids using fuming sulfuric acid is a standard procedure [9] and it apparently applies also to trifluoromethylbenzene sulfonic acid [10]. Thus this route was explored initially to prepare the desired sulfonic acids. In the case of perfluoro-n-heptylbenzene, no reaction took place at 70°C; prolonged exposure at 160°C gave on work-up the sodium salt in ~70% yield. However, the purification was very tedious. The treatment of 1-perfluoro-n-heptylnaphthalene with fuming sulfuric acid at low temperatures, 80°C, resulted mainly in starting material recovery associated with degradation evidenced by black discoloration. At higher temperatures only degradation took place.

In view of the above findings and due to the difficulties envisioned in the subsequent transformations of the sodium salts to other salts, preparation of sulfonyl chlorides was investigated next. Conducting the reaction of chlorosulfonic acid with the perfluoroalkylaromatics at low temperatures resulted in high yields of the respective sulfonyl chlorides as evident from the data listed in Table 2. Based on ^1H NMR analysis, the substitution in the benzene compounds occurred exclusively in the meta position, which would be expected, and in the naphthalenes in the 5 position. The sulfonyl chlorides exhibited mass spectral breakdown patterns characterized by the $R_fArSO_2^+$, R_fAr^+, $CF_2ArSO_2Cl^+$, CF_2Ar^+, and CF_3Ar^+ ions. It should be noted that in the naphthalene compounds the $R_fArSO_2^+$ ion was of relatively low intensity. On the other hand, the rearrangement ion $CF_2C_{10}H_6Cl$ was one of the high intensity peaks.

The sulfonyl chlorides were found to be surprisingly hydrolytically stable; the compounds were recovered unchanged after prolonged (24 h) heating in boiling water. The salts were prepared from the sulfonyl chlorides and the corresponding metal hydroxides. Unfortunately, contrary to expectations the salts, with the possible exception of $[n\text{-}C_8F_{17}(C_6H_4)SO_3]_2Zn$,

TABLE 2

Preparation of perfluoroalkyl-substituted aromatic sulfonyl chlorides

Fluoroaromatic type (mmol)	$ClSO_3H$ (mmol)	Reaction Conditions Init. Temp[a] °C	Time h	Final Temp °C	Time h	R_fArSO_2Cl Yield, %
n-$C_7F_{15}C_6H_5$ (2.24)	15	0	2.5	RT	18	55
n-$C_8F_{17}C_6H_5$ (12.65)	90	0	2.0	RT	44	92
n-$C_7F_{15}C_{10}H_7$ (10.08)	75	-20	1	0	2	84
n-$C_8F_{17}C_{10}H_7$ (13.66)	105	-20	1.5	0	4	74

[a] This is the temperature at which the addition of fluoroaromatic took place; the actual addition was performed usually over a 15-30 min. period.

were essentially insoluble in $Cl(CFClCF_2)_{3-4}Cl$ fluids. It is of interest that $n\text{-}C_8F_{17}(C_6H_4)SO_3Li$ did exhibit some protective action of mild steel under the condition of 'Corrosion Resistance Evaluation Procedure' [11] at <0.01%. Similar results were obtained for $n\text{-}C_8F_{17}(C_{10}H_6)SO_3K$ and $[n\text{-}C_8F_{17}(C_6H_4)SO_3]_2Ba$ salts.

The majority of the salts were found to retain the water of crystallization and some, in particular $[n\text{-}C_8F_{17}(C_6H_4)SO_3]_2Zn$, were actually strongly hygroscopic. As evident from the thermogravimetric analysis (TGA) curves presented in Fig. 1 and 2, with the exception of the aluminum salts of both the benzene and naphthalene sulfonic acids and the naphthalene sulfonic acid potassium salt, $n\text{-}C_8F_{17}(C_{10}H_6)SO_3K$, all the salts appeared to be thermally stable at least to 350°C. The early weight loss observed in some instances followed by leveling off is attributed to the loss of water. In the case of the barium salt of the dinonylnaphthalenesulfonic acid, definite gradual weight loss, with an absence of any leveling off, was observed from 125-350°C (10% weight loss at 350°C) followed by a rapid weight loss at 350°C (~60% weight loss at 400°C). Based on the TGA data it is clearly evident that the presence of perfluorooctyl chain did enhance the thermal stability of the salts as compared to the alkyl-substituted compounds; however, it failed in imparting solubilizing action.

EXPERIMENTAL

All the melting points are uncorrected and were determined in evacuated sealed capillaries. Molecular weights were obtained in hexafluorobenzene with a Mechrolab Model 302 vapor pressure osmometer. Infrared spectra were recorded either neat (on liquids) or as double mulls (Kel-F oil No. 10 and Nujol) using a Perkin-Elmer Corporation infrared spectrophotometer Model 1330. The mass spectra were obtained by direct insertion probe and using combined gas chromatography/mass spectrometry (GC/MS) employing a Du Pont 21-491B double focusing mass

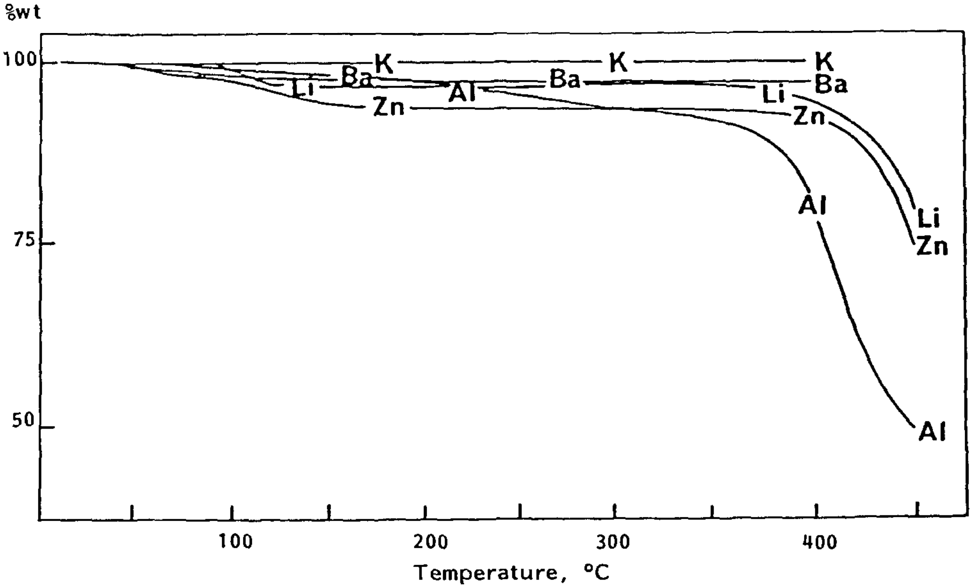


Fig.1. TGA scans of n-$C_8F_{17}(C_6H_4)SO_3M$.

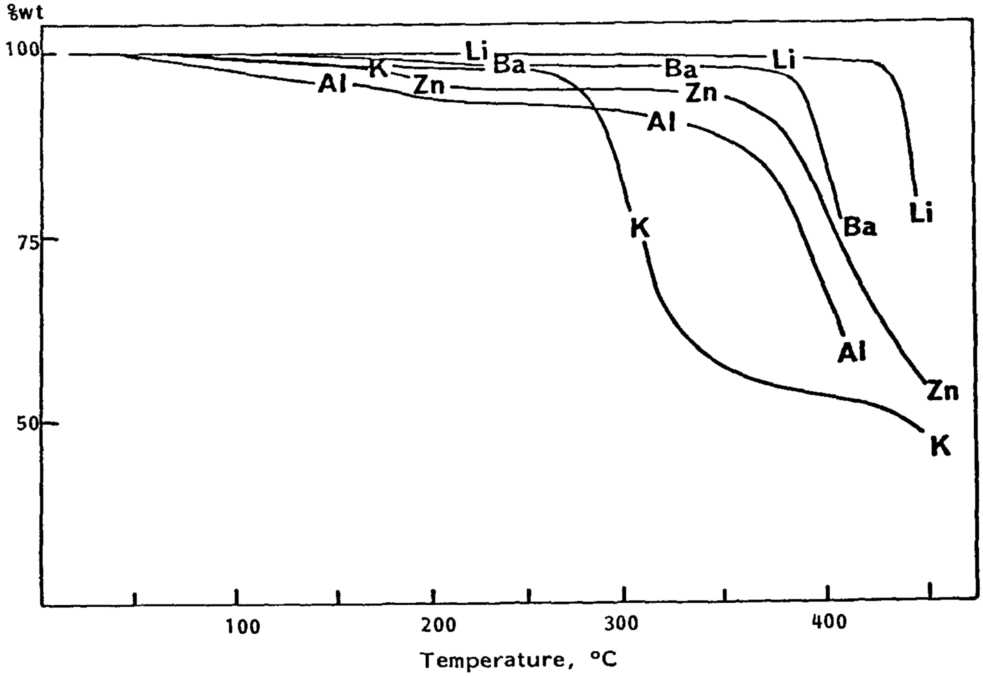


Fig.2. TGA scans of n-$C_8F_{17}(C_{10}H_6)SO_3M$.

spectrometer attached to a Varian Aerograph Model 2700 gas chromatograph (GC) equipped with a flame ionization detector and a Du Pont 21-094 data acquisition and processing system. ^{1}H NMR spectra were recorded on Varian VRX-200 spectrometer in $CDCl_3$; Me_4Si was used as an external standard. Thermogravimetric analyses were carried out in nitrogen from room temperature to 550°C at 10°C/min with a Du Pont 990/951 system. The elemental analyses were performed by Schwarzkopf Microanalytical Laboratory, Woodside, NY.

Coupling Reactions

A mixture of the ingredients as listed in Table 1 (copper bronze, product of BDH Chemicals Ltd., Poole, England was used as received) was heated with stirring at the denoted temperature for the denoted period of time under nitrogen bypass protected from light. Following cooling to room temperature, the products were isolated by addition to a stirred 1:1 mixture of ether and water. The organic layer was subsequently washed several times with water and dried over anhydrous $MgSO_4$. The residue left after solvent removal was either distilled or crystallized.

Preparation of sodium perfluoro-n-heptylbenzenesulfonate

To stirred fuming sulfuric acid (2.64 g) at 150-160°C was added slowly perfluoro-n-heptylbenzene (1.41 g, 3.16 mmol). After 1 h at 160°C, two layers were still present; 17 h exposure afforded a single layer. The material, on pipetting onto ice water (15 mL) followed by salting out with solid sodium chloride (5.9 g), resulted in precipitation of a gray solid (1.47 g, 85% yield) which failed to dissolve on heating. The product was purified by extraction with hot acetonitrile. The material (1.28 g, 74% yield), which crystallized after acetonitrile concentration, exhibited infrared spectrum identical with that of the authentic sample.

Preparation of perfluoroalkyl-substituted arylsulfonyl chlorides

All the reactions were performed in the absence of solvent with rigid exclusion of moisture. To the denoted quantity of cooled (see initial temperature in Table 2) chlorosulfonic acid was added the given quantity of perfluoroalkylaromatic. After the listed interval of time, the temperature was raised to the final value and kept at this temperature for the given period of time. The products were isolated by addition to crushed ice followed by extraction with carbon tetrachloride or carbon tetrachloride/Freon-113 mixture. The organic solution was washed with water, 10% sodium carbonate, and again with water. Following drying over anhydrous $MgSO_4$, the solvent was removed in vacuo and the solid residue was recrystallized from hexane.

Perfluoro-n-octylbenzenesulfonic acid and 1-perfluorooctylnaphthalenesulfonic acid metal salts. Lithium, potassium, barium, zinc, and aluminum salts of the arylsulfonic acids were prepared by treating the arylsulfonyl chlorides at 100°C with a 10% aqueous solution of the given metal hydroxide followed by heating at 100°C for 24 h. All the salts were essentially water insoluble. To remove the inorganic salts, the precipitates were washed with water, and to remove the unreacted sulfonyl chlorides, this was followed by washing with Freon-113. All the salts were dried in vacuo at 100°C.

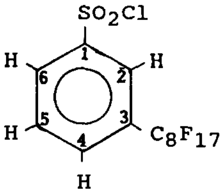
Perfluoro-n-octylbenzene (nc). BP 94-95°C/12 mm Hg. Anal. Calcd for $C_{14}H_5F_{17}$: C, 33.89; H, 1.02; F, 65.10; MW, 496.13. Found: C, 33.85; H, 1.00; F, 65.00; MW, 540. IR (capillary film, cm^{-1}): 3070 (w), 3041 (w), 1454 (m), 1370 (m), 1292 (s), 1245 (s, br), 1210 (s, br), 1113 (s), 1090 (s), 1072 (s), 1050 (m), 1035 (m), 1017 (m), 958 (m), 948 (m), 920 (m), 860 (m), 816 (m), 802 (m), 776 (m), 762 (s), 744 (m), 735 (m), 722 (m), 700 (s), 660 (s), 650 (s). MS (70 eV) m/e (relative intensity, ion): 496 (44.8%, M), 477 (26.0%, M - F), 208 (10.4%, $C_3F_5C_6H_5$), 177 (18.3%, $CF_2CF_2C_6H_5$), 176 (11.8%, $CF_2CF_2C_6H_4$), 158 (28.5%, $C_2F_3C_6H_5$), 131 (16.6%, C_3F_5), 127 (base, $CF_2C_6H_5$), 126 (21.0%, $CF_2C_6H_4$).

1-Perfluoro-n-heptylnaphthalene. MP 39.5-41°C (crystallized from ethanol). Anal. Calcd for $C_{17}H_7F_{15}$: C, 41.15; H, 1.42; F, 57.44; MW, 496.16. Found: C, 40.07; H, 1.37; F, 58.57; MW, 510. IR (Kel-F/Nujol, cm^{-1}): 3060 (w), 1513 (w), 1365 (w), 1350 (w), 1318 (w), 1240 (s), 1203 (s), 1150 (s), 1130 (s), 1100 (m), 1033 (w), 1018 (w), 960 (w), 880 (w), 812 (w), 795 (m), 772 (s), 735 (m), 721 (m), 705 (m), 679 (m), 661 (m). MS (70 eV) m/e (relative intensity, ion): 496 (38.7%, M), 208 (10.0%, $C_2F_3C_{10}H_7$), 177 (base, $CF_2C_{10}H_7$), 157 (10.7%, $CFC_{10}H_6$), 127 (15.4%, $C_{10}H_7$), 69 (19.2%, CF_3).

1-Perfluoro-n-octylnaphthalene. MP 64-65°C. Anal. Calcd for $C_{18}H_7F_{17}$: C, 39.58; H, 1.29; F, 59.13, MW, 546.17. Found: C, 38.02; H, 1.11; F, 60.94; MW, 600. IR (Kel-F/Nujol mull, cm^{-1}): 3055 (w), 1600 (w), 1580 (w), 1515 (m), 1366 (m), 1350 (m), 1325 (m), 1200 (s,br) 1148 (s,br) 1079 (m), 1060 (m), 1043 (m), 1027 (m), 995 (s), 982 (m), 973 (m), 947 (m), 933 (s), 917 (m), 807 (s), 785 (s), 769 (s), 734 (m), 706 (m), 661 (s), 617 (m), 583 (m), 559 (m). MS (70 eV) m/e (relative intensity, ion): 546 (89.1%, M), 527 (31.2%, M - 19), 507 (11.6%, M - 2F - H), 219 (11.2%, C_4F_9), 208 (24.7%, $C_2F_3C_{10}H_7$), 207 (23.5%, $C_2F_3C_{10}H_6$), 177 (base, $CF_2C_{10}H_7$), 175 (23.1%, $CF_2C_{10}H_5$), 169 (12.0%, C_3F_7), 157 (22.6%, $CFC_{10}H_6$), 151 (21.3%, C_3F_6H), 127 (26.6%, $C_{10}H_7$).

m-Perfluoro-n-heptylbenzenesulfonyl chloride (nc). MP 50-51°C. Anal. Calcd for $C_{13}H_4ClF_{15}O_2S$: C, 28.67; H, 0.74; Cl, 6.51; F, 52.32; S, 5.89; MW, 544.68. Found: C, 28.86; H, 0.75; Cl, 6.56; F, 52.25; S, 6.02; MW, 540. IR (Kel-F/Nujol mull, cm^{-1}): 3108 (w), 3080 (w), 1600 (w), 1479 (w), 1427 (m), 1385 (s), 1369 (m), 1320 (m), 1300 (m), 1270 (m), 1247 (m), 1230 (m), 1196 (s), 1147 (s), 1131 (m), 1106 (m), 1073 (m), 995 (m), 942 (w), 907 (w), 860 (w), 811 (w), 775 (w), 747 (w), 708 (m), 678 (m), 658 (m), 597 (w), 577 (w), 560 (w), 550 (w). MS (70 eV) m/e (relative intensity, ion): (only ^{35}Cl ions listed) 544 (28.9%, M), 525 (41.6%, M - F), 509 (base, M - Cl), 493 (23.4%, $C_7F_{15}C_6H_4SO$), 445 (87.6%, $C_7F_{15}C_6H_4$), 395 (23.0%, $C_6F_{13}C_6H_4$), 225 (97.2%, $CF_2C_6H_4SO_2Cl$), 207 (35.7%, $C_3F_5C_6H_4$), 195 (24.5%, $C_2F_5C_6H_4$), 176 (60.0%, $CF_2CF_2C_6H_4$), 145 (64.8%, $CF_3C_6H_4$), 126 (69.1%, $CF_2C_6H_4$), 69 (58.8%, CF_3).

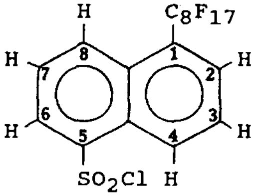
m-Perfluoro-n-octylbenzenesulfonyl chloride (nc). MP 62-64°C. Anal. Calcd for $C_{14}H_4ClF_{17}O_2S$: C, 28.30; H, 0.68; Cl, 5.97; F, 54.36; S, 5.39; MW, 594.68. Found: C, 28.28; H, 1.52; Cl 5.73; F, 54.22; S, 5.33; MW, 620. IR (Kel-F/Nujol mull, cm^{-1}): 1390 (w), 1293 (w), 1240 (s), 1200 (s), 1146 (s), 1115 (m), 1083 (w), 1049 (w), 995 (w), 959 (w), 903 (w), 800 (w), 766 (w), 735 (w), 709 (w), 675 (m), 656 (w), 550 (w). MS (70 eV) m/e (relative intensity, ion): (only ^{35}Cl ions listed) 594 (14.5%, M), 575 (23.1%, M - F), 559 (92.7%, M - Cl), 543 (11.6%, $C_8F_{17}C_6H_4SO$), 495 (48.7%, $C_8F_{17}C_6H_4$), 445 (11.3%, $C_7F_{15}C_6H_4$), 225 (base, $CF_2C_6H_4SO_2Cl$), 195 (18.5%, $C_2F_5C_6H_4$), 176 (41.1%, $CF_2CF_2C_6H_4$), 145 (67.4%, $CF_3C_6H_4$), 126 (51.4%, $CF_2C_6H_4$), 69 (41.5%, CF_3). ^1H NMR: 8.25 (s, H_2), 8.27 (d, H_3, J = 7.2 Hz), 7.96 (d, H_4, J = 7.7 Hz), 7.82 (t, H_5, J = 8.0 Hz) ppm.



1-Perfluoro-n-heptylnaphthalene-5-sulfonyl chloride (nc). MP 99.5-101°C. Anal. Calcd for $C_{17}H_6ClF_{15}O_2S$: C, 34.34; H, 1.02; Cl, 5.96; F, 47.92; S, 5.39; MW, 596.67. Found: C, 34.22; H, 0.99; Cl, 5.74; F, 47.80; S, 5.49; MW, 600. IR (Kel-F/Nujol mull, cm^{-1}): 1510 (w), 1370 (m), 1350 (m), 1340 (m), 1320 (m), 1271 (m), 1238 (s), 1210 (s), 1170 (m), 1142 (s), 1098 (m), 1035 (w), 994 (w), 880 (w), 992 (s), 770 (w), 761 (w), 745 (w), 722 (w), 707 (m), 667 (m), 583 (w). MS (70 eV) m/e (relative intensity, ion): (only ^{35}Cl ions listed) 594 (82.0%, M), 559 (23.2, M - Cl), 495 (57.7%, $C_7F_{15}C_{10}H_6$), 475 (14.7%, $C_7F_{14}C_{10}H_5$), 275 (base, $CF_2C_{10}H_6SO_2Cl$), 226 (60.1%, $CF_2CF_2C_{10}H_6$), 211 (28.6%, $CF_2C_{10}H_6Cl$), 176 (27.1%, $CF_2C_{10}H_6$), 175 (20.9%, $CF_2C_{10}H_5$).

1-Perfluoro-n-octylnaphthalene-5-sulfonyl chloride (nc). MP 103-105°C. Anal. Calcd for $C_{18}H_6ClF_{17}O_2S$: C, 33.54; H, 0.94; Cl 5.50; F, 50.10; S, 4.97; MW, 644.69. Found: C, 33.35;

H, 0.87; Cl, 5.53; F, 50.04; S, 4.98; MW, 650. IR (Kel-F/Nujol mull, cm^{-1}): 1510 (w), 1370 (m), 1350 (w), 1340 (w), 1328 (w), 1289 (m), 1212 (s), 1168 (s), 1146 (s), 1132 (s), 1111 (m), 1095 (w), 1071 (w), 1011 (w), 959 (w), 943 (w), 918 (w), 806 (w), 791 (s), 705 (m), 667 (m), 580 (w). MS (70 eV) m/e (relative intensity, ion): 644 (67.6%, M), 609 (18.9%, M - Cl), 545 (54.7%, $C_8F_{17}C_{10}H_6$), 525 (13.8%, $C_8F_{16}C_{10}H_5$), 275 (base, $CF_2C_{10}H_6SO_2Cl$), 226 (63.7%, $CF_2CF_2C_{10}H_6$), 211 (28.2%, $CF_2C_{10}H_6Cl$), 207 (18.4%, $C_2F_3C_{10}H_6$), 195 (10.3%, $CF_3C_{10}H_6$), 176 (33.0%, $CF_2C_{10}H_6$), 69 (17.3%, CF_3). ^1H NMR: 9.11 (d, H_6, J = 8.7 Hz), 8.62 (d, H_2, J = 8.8 Hz), 8.47 (d, H_4, J = 7.3 Hz), 8.04 (d, H_8, J = 7.32 Hz), 7.90 (t, H_7, J = 8.0 Hz), 7.75 (t, H_3, J = 8.1 Hz) ppm. Assignment based on chemical shift anisotropy of SO_2Cl and selective homonuclear decoupling.



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