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Branched polyfluorinated triflate—An easily available polyfluoroalkylating agent

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Abstract

A novel branched polyfluoroalkyl triflate was prepared from readily available (perfluorohexyl)propyl iodide in five steps with high overall yield. The reactivity of the triflate has been tested in model reactions with methyl gallate (*O*-nucleophile) and benzylamine (*N*-nucleophile). The reactions yielded good amounts of the desired polyfluorinated products.

Keywords: Fluorinated branched triflate; Fluorous building block; Perfluoroalkylating agent; Fluorous tag; Electrophilic fluorinated reagent

1. Introduction

Highly fluorinated compounds (generally called *fluorous* compounds [1–3]) display properties which are utilized in catalysis, organic syntheses, separation techniques, the biomedicinal field, electronics and material chemistry [4]. Substitution of compounds with nucleophilic functional groups (–OH, –SH, $–NH_2$) can be achieved using electrophilic polyfluoroalkylating compounds such as iodides [5–7], acylhalogenides [8,9], anhydrides [10–12], tosylates [13–15], mesylates [13,14], triflates [16–19] or epoxides [20–22].

One of the most reactive classes of polyfluoroalkylating agents are polyfluorinated triflates. Those with ethylene or propylene spacers have been synthesized and utilized successfully for the construction of various highly fluorinated compounds, such as fluorophilic cyclopentadienes [16,17], alkynes [19] or polyfluorinated benzyloxamines [18]. In all cases, using triflates was necessary because other agents (i.e. iodides) failed due to low reactivity. Known polyfluorinated triflates are able to transfer only one polyfluoroalkylated chain. For the connection of two chains, 2 mol of the triflate are necessary. Our aim was to develop a synthetic route for the primary triflate which could transfer two polyfluorinated chains in one step.

We used a known method for the preparation of monoacid with two polyfluorinated chains [23]. In the next step, the carboxylic group was converted into the corresponding alcohol. The final step included a reaction with trifluoromethanesulfonyl anhydride to obtain the desired primary branched polyfluorinated triflate with two polyfluorinated chains. The triflate is sufficiently insulated (five carbon spacer) from the electron withdrawing fluorinated chain in order to preserve its high reactivity.

2. Results and discussion

Our synthetic strategy was based on the substitution of diethyl malonate with two polyfluorinated chains [23]. The diethyl-malonate was deprotonated with NaH and then reacted with 3-(perfluoroalkyl)propyl iodide in anhydrous DMF and diethyl bis(polyfluoroalkyl)malonate **1** was obtained at 98% yield. The basic hydrolysis of polyfluorinated malonate **1** by aqueous–ethanolic potassium hydroxide produced fluorinated dicarboxylic acid **2** (89% yield), which after pyrolysis at 180 °C yielded the corresponding product of decarboxylation, fluorinated monoacid **3** at almost quantitative yield (98%). This acid (**3**) was treated with borane–THF complex in anhydrous THF

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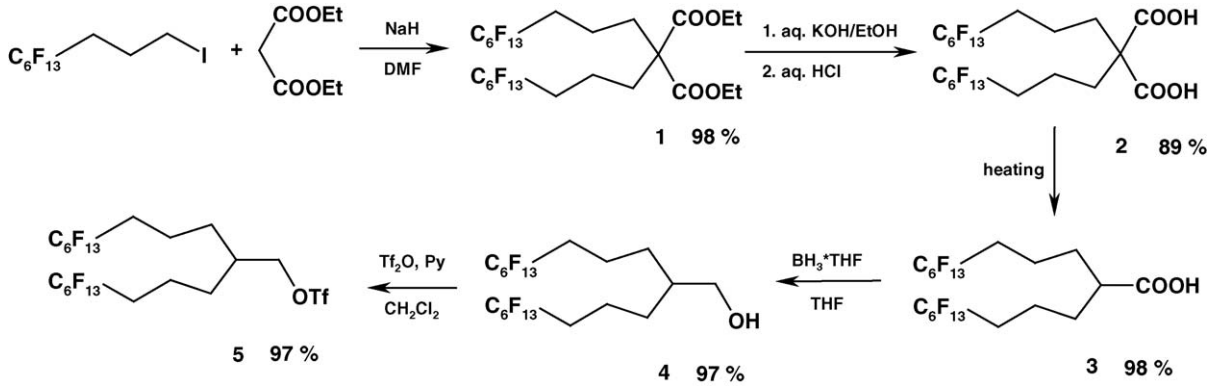
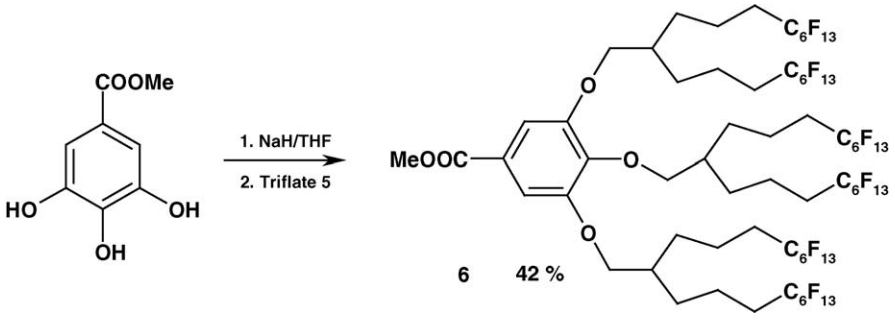
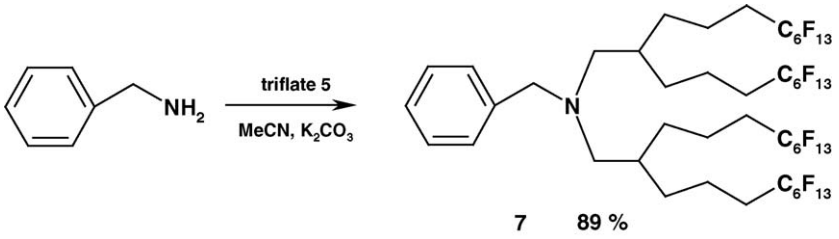


Scheme 1. Preparation of branched polyfluorinated triflate.



Scheme 2. Modification of gallate with fluorinated chains.



Scheme 3. Reaction of benzylamine with triflate **5**.

and 2,2-[bis(polyfluoroalkyl)]ethan-1-ol (**4**) was obtained in high yield (97%). The alcohol **4** was converted into corresponding triflate under same conditions as we used for preparation of [2-(perfluoroalkyl)ethyl]triflates [16]. Thus, the alcohol was reacted with trifluoromethanesulfonic acid anhydride in the presence of pyridine in anhydrous dichloromethane at low temperatures. The desired branched triflate was obtained in excellent yield (97%). The overall yield of the bis(polyfluoroalkylated) triflate **5** was high (80%) (Scheme 1).

To demonstrate the reactivity of our new triflate **5**, we carried out two reactions leading to potentional fluorinated blocks for further building of larger fluorinated units. The modification of gallate with three fluorinated chains in one step was published recently [24–26]. Gallate was transformed into the corresponding sodium salt which was reacted with triflate **5** under reflux. The reaction afforded the corresponding gallate **6** substituted with six fluorinated chains (Scheme 2).

The fluorinated gallate **6** could be used as a potential precursor for the construction of larger fluorous tags [23].

We tested the reactivity of a primary amino group toward the triflate **5**. We chose a reactive amine—benzylamine. By means of triflate **5**, we connected four fluorinated chains in one step. Benzylamine bearing only two fluorinated chains was synthesized by this point [27].

The reaction was carried out in acetonitrile with potassium carbonate as a base. The triflate was used in excess (2.5 equivalents versus amino group). The mixture was heated to 80 °C for 5 h and produced the corresponding tertiary amine **7** in yield 89% (Scheme 3).

The gallate **6** (61.6% F) and amine **7** (61.0% F) can be regarded as fluorophilic compounds, because their content of fluorine is over 60% [28].

3. Conclusions

We developed a facile method for the preparation of branched triflate from commercially available compounds with an overall yield of 80%. Its reactivity was tested in

reactions with benzylamine and methyl gallate. This triflate was found to be a suitable polyfluoroalkylating agent for construction of highly fluorinated structures (e.g. fluorous tags or dendrones).

4. Experimental

4.1. General

NMR spectra were recorded on a Varian Gemini 300 HC (FT, ^{1}H at 300 MHz, ^{13}C at 75 MHz, ^{19}F at 281 MHz) instrument using TMS and $CFCl_3$ as the internal standards. Chemical shifts are quoted in ppm (δ, scale; s, singlet; bs, broad singlet; d, doublet; t, triplet; q, quadruplet; m, multiplet), coupling constants J in Hertz, solvents $CDCl_3$, CD_3OD. Mass spectrometry analyses were performed on a Hewlett–Packard GC HP5890 instrument. IR spectrometry analyses were performed on a Bruker IFS 66v/S instrument.

The chemicals used were as follows: benzylamine, diethyl malonate, 3-(perfluorohexyl)propyl iodide, sodium hydride (60% suspension in oil), trifluoromethanesulfonic acid anhydride, methyl gallate (all Aldrich). Silica gel (60–100 μm, Merck). Anhydrous dichloromethane, pyridine and *N*,*N*-dimethylformamide were purchased from Fluka. Other solvents were purchased from Penta and dried according to standard procedures.

4.2. Preparation of diester **1**

To stirred suspension of NaH (60% suspension in oil; 270 mg, 6.75 mmol; washed with petroleum ether before use) in anhydrous DMF (15 mL) under argon atmosphere the solution of diethyl malonate (360 mg, 2.25 mmol) in anhydrous DMF (7 mL) was added. After stirring for 1 h at room temperature, 3-(perfluorohexyl)propyl iodide (2.490 g, 5.1 mmol) in anhydrous DMF (7 mL) was added dropwise. The mixture was heated to 80 °C and stirred for 12 h. After cooling, water (150 mL) was added and the mixture was extracted with diethyl ether (3 × 150 mL). The organic fraction was washed with brine (3 × 100 mL) and dried over Na_2SO_4. Diethylether was removed under reduced pressure. The crude product was obtained as a brown oil (1.943 g, 98%) and used for the next step without further purification.

^{1}H NMR ($CDCl_3$) δ 1.25 (t, 6H, $^3J_{HH}$ = 7.0 Hz), 1.55 (m, 4H), 1.97 (t, 4H, $^3J_{HH}$ = 8.5 Hz), 2.19 (m, 4H), 4.21 (q, 4H, $^3J_{HH}$ = 7.0 Hz); ^{13}C NMR ($CDCl_3$) δ 13.9 (s, 2C), 15.5 (s, 2C), 31.0 (t, 2C, $^2J_{CF}$ = 21 Hz), 32.2 (s, 2C), 57.2 (s, 1C), 61.5 (s, 2C), 108.5–121.2 (m, 12C), 170.8 (s, 2C); ^{19}F NMR ($CDCl_3$) δ −81.8 (t, 6F, $^3J_{FF}$ = 10.4 Hz), −115.2 (m, 4F), −123.0 (m, 4F), −124.0 (m, 4F), −124.7 (m, 4F), −127.3 (m, 4F).
IR ($CDCl_3$) (cm^{-1}) 1727, 1243, 1237, 1207, 1146.
MS: for $C_{25}H_{22}F_{26}O_4$ calculated: 880, found: (M) 880, (M-H) 879.
Anal. calcd. for $C_{25}H_{22}F_{26}O_4$: C, 34.11; H, 2.52. Found: C, 33.93; H, 2.53.

4.3. Preparation of diacid **2**

The diester **1** (3.525 g, 4 mmol) was dissolved in 96% ethanol (25 mL) and KOH (50% aqueous solution, 25 mL) was added. The reaction mixture was heated at 80 °C overnight. After cooling, the reaction mixture was acidified with 36% aqueous solution HCl (pH ∼ 2). A brown precipitate was filtered off, washed with water (250 mL), dissolved in ethyl acetate and dried over Na_2SO_4. Ethyl acetate was removed under reduced pressure and crude diacid **2** was obtained (3.167 g) as a brown solid. Crude product was recrystallized from hot toluene to give **2** (2.276 g, 69%) as a pale yellow powder. The next crop of **2** (0.654 g, 20%) was obtained by concentration of mother liquor.

^{1}H NMR (CD_3OD) δ 1.62 (m, 4H), 2.05 (m, 4H), 2.31 (tt, 4H, $^3J_{HH}$ = 7.6 Hz, $^3J_{HF}$ = 19.6 Hz); ^{13}C NMR (CD_3OD) δ 16.8 (s, 2C), 31.9 (t, 2C, $^2J_{CF}$ = 22 Hz), 33.4 (s, 2C), 58.3 (s, 1C), 174.6 (s, 2C); ^{19}F NMR (CD_3OD) δ −81.7 (t, 6F, $^3J_{FF}$ = 10 Hz), −114.5 (m, 4F), −122.5 (m, 4F), −123.4 (m, 4F), −124.1 (m, 4F), −126.8 (m, 4F).
IR (KBr) (cm^{-1}) 3440, 1704, 1237, 1209, 1143.
MS: for $C_{21}H_{14}F_{26}O_4$ calculated: 824, found: (M-H) 823; mp = 139–141 °C.
Anal. calcd. for $C_{21}H_{14}F_{26}O_4$: C, 30.60; H, 1.71. Found: C, 30.18; H, 1.72.

4.4. Preparation of monoacid **3**

The diacid **2** (recrystallized, 884 mg, 1.07 mmol) was heated at 190 °C for 45 min. After cooling, the crude monoacid was dissolved in chloroform and filtered (diacid **2** is almost insoluble in $CHCl_3$). Chloroform was removed under reduced pressure and light brown waxy product was obtained (815 mg, 98%).

^{1}H NMR ($CDCl_3$) δ 1.69 (m, 8H), 2.09 (m, 4H), 2.45 (m, 1H); ^{13}C NMR ($CDCl_3$) δ 18.1 (s, 2C), 30.6 (t, 2C, $^2J_{CF}$ = 21 Hz), 31.3 (s, 2C), 44.9 (s, 1C), 108.5–121.2 (m, 12C), 181.3 (s, 1C); ^{19}F NMR ($CDCl_3$) δ −81.5 (t, 6F, $^3J_{FF}$ = 10 Hz), −114.9 (m, 4F), −122.5 (m, 4F), −123.5 (m, 4F), −124.1 (m, 4F), −126.8 (m, 4F).
IR ($CDCl_3$) (cm^{-1}) 1709, 1241, 1210, 1145.
MS: for $C_{20}H_{14}F_{26}O_2$ calculated: 780, found: 780, (M-H) 779; mp = 44–45 °C.
Anal. calcd. for $C_{20}H_{14}F_{26}O_2$: C, 30.79; H, 1.81. Found: C, 30.80; H, 1.85.

4.5. Preparation of alcohol **4**

The monoacid **3** (810 mg, 1.04 mmol) was dissolved in anhydrous THF (2 mL). Under argon atmosphere, borane–THF complex (1 M solution in THF, 5.2 mL, 5.2 mmol) was added dropwise. Reaction mixture was refluxed for 16 h. After cooling down, 20% aqueous solution HCl (10 mL) was carefully added to destroy excess of borane. Water (100 mL) was added and mixture was extracted with diethyl ether

(3 × 150 mL). Organic fractions were washed with water (2 × 100 mL) and brine (100 mL) and dried over Na_2SO_4. Diethylether was removed under reduced pressure and product was obtained as a white waxy solid (771 mg, 97%).

^{1}H NMR ($CDCl_3$) δ 1.40 (m, 5H), 1.61 (m, 4H), 2.03 (tt, 4H, $^3J_{HH}$ = 7.3 Hz, $^3J_{HF}$ = 18.5 Hz), 2.69 (bs, 1H), 3.55 (d, 2H, $^3J_{HH}$ = 4.4 Hz); ^{13}C NMR ($CDCl_3$) δ 17.6 (s, 2C), 30.4 (s, 2C), 31.1 (t, 2C, $^2J_{CF}$ = 21 Hz), 40.2 (s, 1C), 64.4 (s, 1C), 108.5–121.2 (m, 12C); ^{19}F NMR ($CDCl_3$) δ −82.8 (t, 6F, $^3J_{FF}$ = 10.4 Hz), −116.1 (m, 4F), −123.6 (m, 4F), −124.6 (m, 4F), −125.3 (m, 4F), −128.0 (m, 4F).
IR ($CDCl_3$) (cm^{-1}) 1241, 1215, 1145.
MS: for $C_{20}H_{16}F_{26}O$ calculated: 766, found: (M-HF) 746; mp = 48–49 °C.
Anal. calcd. for $C_{20}H_{16}F_{26}O$: C, 31.35; H, 2.10. Found: C, 31.14; H, 2.18.

4.6. Preparation of triflate 5

The solution of trifluoromethanesulfonic acid anhydride (723 mg, 2.56 mmol) in anhydrous dichloromethane (20 mL) was cooled to −15 °C in an ethanol/dry ice bath. Under argon atmosphere, the solution of alcohol **4** (1310 mg, 1.71 mmol) and pyridine (135 mg, 1.71 mmol) in anhydrous dichloromethane (15 mL) was slowly added while stirring. Then the mixture was slowly warmed to room temperature. The reaction mixture was evaporated to dryness and dark oily residue was purified by column chromatography on silica gel (eluent:dichloromethane). The solvent was removed under reduced pressure and triflate **5** was obtained as a light brown oil (1490 mg, 97%).

^{1}H NMR ($CDCl_3$) δ 1.51 (m, 4H), 1.65 (m, 4H), 1.87 (m, 1H), 2.10 (heptet, 4H, $^3J_{HH}$ = 7.0 Hz, $^3J_{HF}$ = 18.2 Hz), 4.49 (d, 2H, $^3J_{HH}$ = 5.0 Hz); ^{13}C NMR ($CDCl_3$) δ 17.5 (s, 2C), 29.7 (s, 2C), 30.9 (t, 2C, $^2J_{CF}$ = 21 Hz), 38.1 (s, 1C), 78.2 (s, 1C), 108.5–121.2 (m, 12C), 118.0 (q, $^1J_{CF}$ = 317 Hz; ^{19}F NMR ($CDCl_3$) δ −75.1 (s, 3F), −81.4 (t, 6F, $^3J_{FF}$ = 10 Hz), 114.8 (m, 4F), −122.4 (m, 4F), −123.4 (m, 4F), −124.1 (m, 4F), −126.8 (m, 4F).
IR (neat) (cm^{-1}) 2961, 1418, 1245, 1207, 1145.
MS: for $C_{21}H_{15}F_{29}O_3S$ calculated: 898, found: (M-CF_3SO_3) 749.
Anal. calcd. for $C_{21}H_{15}F_{29}O_3S$: C, 28.08; H, 1.68. Found: C, 27.97; H, 1.68.

4.7. Substitution of gallate

A mixture of sodium hydride (7.8 mg, 0.33 mmol), methyl gallate (20 mg, 0.11 mmol) and THF (2 mL) was stirred under argon at room temperature for 30 min. The solution of triflate **5** (430 mg, 0.48 mmol) in THF (2 mL) was added and the reaction mixture was heated to 60 °C for 2 h. After cooling, water (20 mL) was added and the mixture extracted with dichloromethane (2 × 20 mL). Organic fractions were dried over $MgSO_4$. Dichloromethane was removed under reduced pressure and, the oily residue was purified by column chromatography on silica gel (eluent:hexan/dichloromethane, 1:1). Product **6** was obtained as a pale yellow oil (112 mg, 42%).

^{1}H NMR ($CDCl_3$) δ 1.66 (m, 24H), 1.88 (m, 3H), 2.06 (m, 12H), 3.90 (s, 3H), 3.91 (d, 2H, $^3J_{HH}$ = 4.7 Hz) 3.96 (d, 4H, $^3J_{HH}$ = 4.7 Hz), 7.28 (s, 2H); ^{13}C NMR ($CDCl_3$) δ 17.7 (s, 6C), 29.7 (s, 6C), 30.9 (t, 6C, $^2J_{CF}$ = 21 Hz), 38.2 (s, 2C), 39.0 (s, 1C), 52.2 (s, 1C), 70.8 (s, 2C), 75.1 (s, 1C), 104.0–120.2 (m, 36C), 107.8 (s, 2C), 125.3 (s, 1C), 141.8 (s, 1C), 152.5 (s, 2C), 166.6 (s, 1C); ^{19}F NMR ($CDCl_3$) δ −81.7 (m,18F), −115.1 (m, 12F), −122.6 (m, 12F), −123.6 (m, 12F), −124.3 (m, 12F), −126.9 (m, 12F).
IR ($CDCl_3$) (cm^{-1}) 2927, 1716, 1241, 1209, 1145.
MS: for $C_{68}H_{50}F_{78}O_5$ (MH) calculated: 2429, found: 2429.
Anal. calcd. for $C_{68}H_{50}F_{78}O_5$: C, 33.62; H, 2.07. Found: C, 33.65; H, 2.08.

4.8. Substitution of benzylamine

A mixture of triflate **5** (100 mg, 0.11 mmol), benzylamine (6 mg, 0.056 mmol), anhydrous potassium carbonate (17 mg, 0.12 mmol) and acetonitrile (2 mL) was stirred under argon at 80 °C for 5 h. After cooling, water (10 mL) was added and the mixture was extracted with chloroform (2 × 50 mL). Organic fractions were dried over $MgSO_4$. Chloroform was removed under reduced pressure and the oily residue was purified by column chromatography on silica gel (eluent:dichloromethane). Product **7** was obtained as a viscous pale yellow oil (80 mg, 89%).

^{1}H NMR ($CDCl_3$) δ 1.20–1.60 (m, 18H), 2.05 (m, 8H), 2.23 (d, 4H, $^3J_{HH}$ = 6.9 Hz), 3.48 (s, 2H), 7.26 (m, 5H); ^{13}C NMR ($CDCl_3$) δ 17.2 (s, 4C), 31.1 (t, 4C, $^2J_{CF}$ = 21 Hz), 31.5 (s, 4C), 35.9 (s, 2C), 59.7 (s, 2C), 60.2 (s, 1C), 104.0–122 (m, 24C), 127.2 (s, 1C), 128.2 (s, 2C), 129.1 (s, 2C), 139.4 (s, 1C); ^{19}F NMR ($CDCl_3$) δ −81.5 (m, 12F, $^3J_{FF}$ = 10 Hz), −114.9 (m, 8F), −122.5 (m, 8F), −123.4 (m, 8F), −124.2 (m, 8F), −126.9 (m, 8F).
IR ($CDCl_3$) (cm^{-1}) 2857, 1243, 1192, 1145.
MS: for $C_{47}H_{37}F_{52}N$ (MH) calculated: 1603, found: (M-H) 1602.
Anal. calcd. for $C_{47}H_{37}F_{52}N$: C, 35.20; H, 2.33. Found: C, 35.07; H, 2.39.

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