(m, 2 H, $CH_2C{=}N$), 4.18-4.23 (m, 2 H, CH_2N), 6.19 (dd, J = 4, 3 Hz, 1 H, pyrrole), 6.73 (dd, J = 3, 1 Hz, 1 H, pyrrole), 7.07-7.13 (m, 1 H, Ph), 7.19 (dd, J = 4, 1 Hz, 1 H, pyrrole), 7.30, 7.35 (m, 2 H, Ph), 7.49-7.54 (m, 2 H, Ph), 8.25 (s, 1 H, NH); ^{13}C NMR δ 24.26, 27.56 ($CH_2CH_2CH_2N$), 32.63 ($CH_2C{=}N$), 49.56 (CH_2N), 108.56, 119.53, 119.92, 123.93, 127.09, 129.03, 137.39 (pyrrole + Ph), 152.36 (C=N), 153.90 (C=O).

Furoxans 9 from Reaction of Nitro Heterocycles 5a, 5d, and 13a with Phenyl Isocyanate-Triethylamine. 9a from **5a** (yield 35%, oil): ^1H NMR δ 2.10 (t, J = 7 Hz, 2 H, $CH_2C{=}N$), 2.25 (m, 2 H, $CH_2C{=}N{\rightarrow}O$), 4.02 (t, J = 7 Hz, 2 H, CH_2N), 4.18 (m, CH_2N), 6.51 (d, J = 3 Hz, 2 H, Ind.), 6.66 and 6.68 (d, J = 3 Hz, 1 H, Ind.), 7.20-7.40 (m, 6 H, Ind.), 7.57-7.62 (m, 2 H, Ind.).

9d from **5d** (yield 42%, oil): δ 1.80-2.00 (m, 4 H, CH_2CH_2N), 2.03 (t, J = 7 Hz, 2 H, $CH_2C{=}N$), 2.15 (t, J = 7 Hz, 2 H, CH_2N), 4.62 (t, J = 7 Hz, 2 H, CH_2N), 6.24 (s, 2 H, Ind.), 7.05-7.52 (m, 8 H, Ind.).

9e from 13a (yield 32%, oil): δ 2.40 and 2.52 (t, J = 7 Hz, 2 H, $CH_2C{=}N$), 3.92 and 3.96 (t, J = 7 Hz, 2 H, CH_2N), 6.12 and 6.60 (m, 4 H, pyrrole).

Preparation of 3,3-Difluoroacrylic Acid Derivatives by Dehydrohalogenation of Activated Acyl Compounds. Synthesis of Two Potential Precursors to Difluoropropadienone

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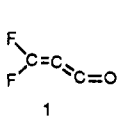
We have recently reported the successful preparation of difluoropropadienone (**1**) by flash vacuum pyrolysis of 3,3-difluoroacrylic anhydride.[1] Attempts to prepare this anhydride directly from 3,3-difluoroacrylic acid[2] were unsuccessful under a variety of conditions and invariably led to destruction of the starting material, presumably due to conjugate addition and polymerization.

Due to the difficulty in obtaining 3,3-difluoroacrylic anhydride, other possible methods for making difluoropropadienone were explored. One of these routes was the zinc reduction of 2-bromo-3,3-difluoroacryloyl chloride (**2**). Zinc reduction of 2,2-dibromomalonyl chloride has been used to prepare carbon suboxide in good yield.[3] More recently, alkyl- and arylpropadienones have been trapped in solution by the in situ reduction of 2-bromoacryloyl chlorides using $Mn(CO)_5^-$.[4]



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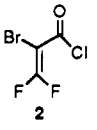
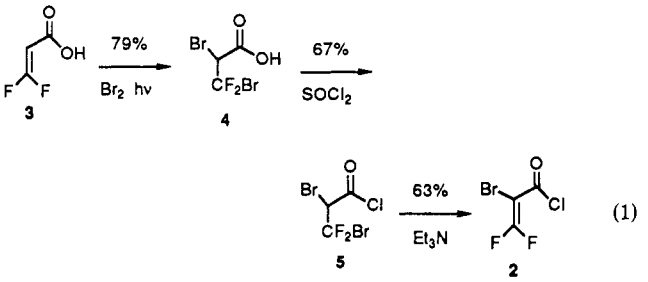


2

In our efforts to synthesize **2** as a precursor to difluoropropadienone, we discovered that it was possible to prepare this, as well as several other 3,3-difluoroacrylic acid derivatives, including 3,3-difluoroacrylic anhydride, by dehydrohalogenation of 3-bromo-3,3-difluoropropionate derivatives. While this elimination reaction has been reported for the preparation of ethyl 3,3-difluoroacrylate from ethyl 3-bromo-3,3-difluoropropionate,[5] there are no reports of this reaction being attempted on activated acyl derivatives.

Results and Discussion

It was possible to prepare 2-bromo-3,3-difluoroacryloyl chloride (**2**) in good yield by the route shown in eq 1.

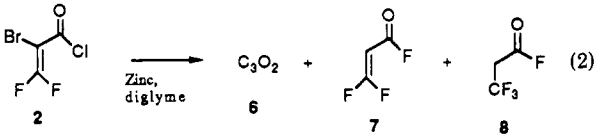


(1)

Photochemical addition of bromine to 3,3-difluoroacrylic acid (**3**)[2] gave **4**, which could be converted to the corresponding acid chloride **5** by heating with excess thionyl chloride. Acid chloride **5** can be dehydrohalogenated to give **2** in 63% yield by treatment with slightly less than 1 equiv of triethylamine in dichloromethane at 0 °C under rigorously anhydrous conditions.

Dehydrohalogenation of an acid chloride with a tertiary amine is a standard method for the formation of a ketene, presumably via an acylammonium intermediate,[6] and this pathway was considered as a potentially serious side reaction. Apparently the increased acidity of the proton adjacent to the two neighboring fluorine atoms together with the leaving group ability of bromine favors elimination to give the desired product rather than the undesired ketene.

With **2** in hand, we were in a position to investigate the zinc reduction reaction. Treatment of **2** with activated zinc dust in the absence of solvent resulted in loss of starting material without the formation of any new volatile products. Addition of **2** to a mixture of activated zinc dust in diglyme at 80 °C resulted in the formation of carbon suboxide (**6**) and 3,3-difluoroacryloyl fluoride (**7**) along with a small amount of 3,3,3-trifluoropropionyl fluoride (**8**) in about a 10% combined yield (eq 2).



(2)

These compounds have been reported in the literature,[7] but we prepared them by independent synthesis for comparison with the reaction product mixture. Carbon suboxide was prepared by treatment of malonic acid with P_2O_5.[8] Compounds **7** and **8** were prepared by the routes shown in eqs 3 and 4.

3-Bromo-3,3-difluoropropionic acid[9] (**9**) was converted to acid chloride **10**, which in turn was converted to the corresponding acid fluoride **11** with use of neat HF-

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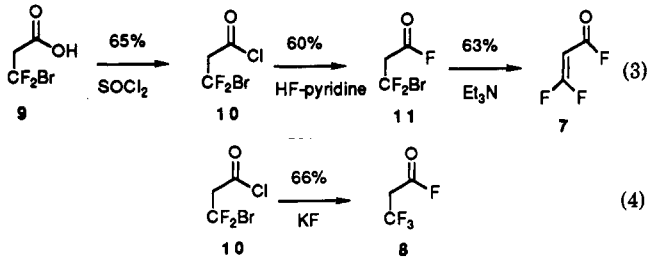
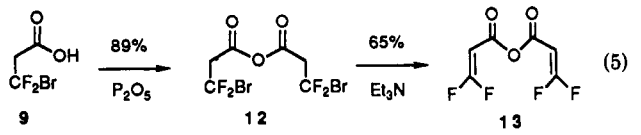


pyridine.[10] This acid fluoride can be dehydrohalogenated by treatment with triethylamine in *o*-dichlorobenzene. The acid fluoride **8** can be prepared simply by treating acid chloride **10** with anhydrous KF in *o*-dichlorobenzene followed by fractionation.[11]

Since the dehydrohalogenative method allowed the preparation of a number of different 3,3-difluoroacryloyl compounds, we applied this procedure to the synthesis of 3,3-difluoroacrylic anhydride (**13**), our original objective. Thus we were eventually able to synthesize 3,3-difluoroacrylic anhydride by the route shown in eq 5.



Acid **9** was converted in high yield to anhydride **12** by using P_2O_5. Compound **12** was treated with slightly less than 2 equiv of triethylamine in methylene chloride at 0 °C to give pure **13** as a white crystalline solid in 65% yield. As reported previously, flash vacuum pyrolysis of **13** produces difluoropropadienone in high yield and purity.[1]

In conclusion, the dehydrohalogenation of 3-bromo-3,3-difluoropropionyl compounds is a reliable method for the preparation of a number of 3,3-difluoroacryloyl derivatives.

Experimental Section

General. All reactions were performed under a dry nitrogen atmosphere unless specified. All glassware was flame dried under high vacuum before use. Tetrahydrofuran and diethyl ether were freshly distilled from sodium/benzophenone ketyl. Dichloromethane was distilled from calcium hydride. Triethylamine was distilled from calcium hydride and stored over freshly activated 4-Å molecular sieves under dry nitrogen prior to use. Proton NMR spectra were observed at 250 MHz and ^{19}F NMR spectra were observed at 188 MHz. All NMR spectra were taken in $CDCl_3$. ^1H NMR chemical shifts are reported with reference to $CHCl_3$ at 7.24 ppm. ^{19}F NMR chemical shifts are reported as ppm upfield from $CFCl_3$ as internal standard. IR spectra were recorded as solutions in $CHCl_3$ unless otherwise noted. Preparative GLC separations were carried out on a 12 ft × $^1/_4$ in. column packed with 10% SF-96 on Chromosorb-P. Dynamic vacuum transfer refers to fractionation through a series of U-bulbs cooled to different temperatures at a pressure of 1 mTorr.

2,3-Dibromo-3,3-difluoropropionic Acid (4). 3,3-Difluoroacrylic acid[2] (2.1 g, 19.5 mmol) was dissolved in ether (7 mL), and a solution of bromine (3.2 g, 20 mmol) in carbon tetrachloride (32 mL) was added. The mixture was irradiated with a 175-W incadescent bulb for 15 min. Solvent was removed under reduced pressure, and the crude product was dissolved in ether (40 mL). The solution was extracted with saturated $NaHCO_3$ (3 × 30 mL). The bicarbonate extract was acidified to pH = 1.5 with concentrated HCl and extracted with 3 × 30 mL of ether. The ether extract was dried over $MgSO_4$ and filtered, and the solvent was removed under reduced pressure to yield 4.19 g (79%) of a white solid, which was used without further purification. An analytical sample of **4** was further purified by sublimation at 1 mTorr to give a white solid, mp 49–51 °C. IR: 3200–2900, 1740 cm^{-1}. ^1H NMR: δ 4.86 (dd, J = 51.5, 14.16 Hz, 1 H), 10.32 (s, 1 H). ^{19}F NMR: δ 49.8 (dd, J = 5.08, 161.2 Hz, 1 F), 55.2 (dd, J = 14.4, 161.2 Hz, 1 F). Anal. Calcd: C, 13.5; H, 0.75. Found: C, 13.45; H. 0.76.

2,3-Dibromo-3,3-difluoropropionyl Chloride (5). 2,3-Dibromo-3,3-difluoropropionic acid (**4**) (3.32 g, 12.4 mmol) and thionyl chloride (17.8 g, 147 mmol) were heated to reflux for 6 h, and the product was purified by vacuum distillation to yield 2.38 g (67%) of a colorless liquid, bp 68–70 °C (35 mmHg). IR: 1800 cm^{-1}. ^1H NMR: δ 5.12 (dd, J = 5.12, 13.1 Hz 1 H). ^{19}F NMR: δ 50.1 (dd, J. = 5.2, 163.7 Hz, 1 F), 55.3 (dd, J = 12.9, 163.6 Hz, 1 F). HR-MS: (M + H^+) 284.8141, calcd for $C_3H_3{}^{79}Br_2{}^{35}ClF_2O$ 284.8129. Anal. Calcd: C, 12.58; H, 0.35. Found: C, 12.78; H, 0.40.

2-Bromo-3,3-difluoroacryloyl Chloride (2). 2,3-Dibromo-3,3-difluoropropionyl chloride (3.36 g, 12.5 mmol) in dichloromethane (25 mL) was cooled for 15 min on an ice–water bath. Triethylamine (1.57 mL, 11.25 mmol) was added over the course of 1 min, and the mixture was stirred for 2 min at 0 °C. The volatile material was removed from the salts by vacuum transfer, and the product was purified by distillation to yield 1.62 g (63%) of a colorless liquid, bp 101 °C. IR (thin film): 1770, 1690 cm^{-1}. ^{19}F NMR: δ 53.4 (d, J = 39.1 Hz, 1 F), 55.3 ppm (d, J = 39.1 Hz, 1 F). HR-MS: (M^+) 205.8764, calcd for $C_3F_2{}^{81}Br{}^{35}ClO$ 205.8769.

3-Bromo-3,3-difluoropropionyl Chloride (10). 3-Bromo-3,3-difluoropropionic acid (**9**) (2.63 g, 13.3 mmol) and thionyl chloride (6.32 g, 46 mmol) were refluxed for 6 h. The product was purified by distillation at atmospheric pressure to yield 1.80 g (65%) of a colorless liquid, bp 117 °C. An analytical sample of **10** was purified by preparative gas chromatography. IR: 1820, 1130, 1030 cm^{-1}. ^1H NMR: δ 3.99 (t, J = 12.3 Hz, 2 H). ^{19}F NMR: δ 46.8 (t, J = 12.3 Hz, 2 F). HR-MS: (M + H^+) 206.900, calcd for $C_3H_2F_2{}^{79}Br{}^{35}ClO$ 206.902. Anal. Calcd: C, 17.35; H, 0.96. Found: C, 17.31; H, 0.94.

3-Bromo-3,3-difluoropropionyl Fluoride (11). 3-Bromo-3,3-difluoropropionyl chloride (**10**) (1.62 g, 7.83 mmol) and 90% HF–pyridine (1.2 mL) were combined in a 10-mL polyethylene vial equipped with a loose fitting cap and magnetic stirrer. The mixture was stirred at room temperature for 21 h. The mixture was poured into anhydrous KF (2 g) and *o*-dichlorobenzene (3 mL). The mixture was shaken vigorously for 1 min and filtered through sand. The product was isolated by vacuum transfer through a series of two traps, one at −20 °C and one at −78 °C. The product was isolated in the −78 °C trap to yield 900 mg (60%) of a colorless liquid, bp 88 °C. An analytical sample of **11** was purified by preparative gas chromatography. IR: 1870, 1220, 1150 cm^{-1}. ^1H NMR: δ 3.68 (t, J = 12.1 Hz, 2 H). ^{19}F NMR: −49.2 (t, J = 11.5 Hz, 1 F), 46.3 ppm (q, J = 11.8 Hz, 2 F). HR-MS: (M + H^+) 190.9318, calcd for $C_3H_3{}^{79}BF_3O$ 190.9319.

3,3-Difluoroacryloyl Fluoride (7). 3-Bromo-3,3-difluoropropionyl fluoride (**11**, 700 mg, 3.66 mmol) was dissolved in *o*-dichlorobenzene (3 mL) and was stirred and cooled in an ice bath for 15 min. Triethylamine (0.51 mL, 3.65 mmol) was added dropwise over the course of 2 min. The product was isolated by dynamic vacuum transfer through a series of two traps, one at −15 °C and one at −196 °C. Pure product was isolated in the −196 °C trap to yield 310 mg (77%) of colorless liquid, bp 45 °C. IR: 1840, 1720 cm^{-1}. ^1H NMR: δ 5.05 (ddd, J = 1.16, 4.29, 20.6 Hz, 1 H). ^{19}F NMR: δ −40.2 (ddd, J = 4.3, 31.8, 33.5 Hz, 1 F), 55.8 (ddd, J = 20.5, 31.8, 37.7 Hz, 1 F), 60.7 ppm (ddd, J = 1.0, 33.6, 37.7 Hz, 1F). HR-MS: (M^+) 109.9987, calcd for C_3HF_3O 109.9980.

3,3,3-Trifluoropropionyl Fluoride (8). 3-Bromo-3,3-difluoropropionyl chloride (**10**) (853 mg, 4.2 mmol) was dissolved in tetraethylene glycol dimethyl ether under nitrogen. Potassium fluoride (318 mg, 5.3 mmol, dried under high vacuum at 250 °C for 24 h) was added, and the mixture was allowed to stir at room temperature for 18 h. Pure product was isolated by vacuum transfer to a −196 °C U-trap to give 260 mg (66%) of colorless liquid. An analytical sample of **8** was purified by preparative GLC. IR (thin film): 1870, 1300, 1200 cm^{-1}. ^1H NMR: δ 3.4 (q, J = 9.0 Hz, 2 H). ^{19}F NMR: δ −49.3 (q, J = 11.5 Hz, 1 F), 64.05 (dt, J = 11.5, 9.2 Hz, 3 F). HR-MS (M^+) 130.0031, calcd for $C_3H_2F_4O$ 130.0042.

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3-Bromo-3,3-difluoropropionic Anhydride (12). Phosphorus pentoxide (4.21 g, 29.6 mmol) and 3-bromo-3,3-difluoropropionic acid (9) (4.59 g, 24.3 mmol) were heated to 70 °C for 3 h under N_2. The reaction flask was equipped with a short-path distillation head, and product was distilled out of the reaction mixture to yield 3.9 g (89%) of a colorless oil, bp 80 °C (0.5 mmHg). This was used without further purification. IR (thin film): 1830, 1200–1050 cm^{-1}. ^1H NMR: δ 3.74 (t, J = 12.1 Hz, 2 H). ^{19}F NMR: δ 45.3 (t, J = 12.6 Hz, 2 F).

3,3-Difluoroacrylic Anhydride. 3-Bromo-3,3-difluoropropionic anhydride (12, 773 mg, 2.15 mmol) was dissolved in dichloromethane (5 mL) under N_2, and the mixture was cooled in an ice–water bath for 10 min. Triethylamine (480 μL, 3.4 mmol) was added by syringe over the course of 2 min with rapid stirring. The mixture was allowed to stir for 1 min a 0 °C. The flask was equipped with a vacuum adaptor, and the product was purified by dynamic vacuum transfer through a series of two traps, one at −20 °C and one at −196 °C. The product was collected as a white crystalline solid in the −20 °C trap and was further purified by a second fractionation to give 210 mg (65%) of a white solid, mp 42–42.5 °C. IR: (gas phase) 1760, 1720 cm^{-1}. ^1H NMR: δ 5.05 (dd J = 1.5, 20.2 Hz, 2 H). ^{19}F NMR: δ 58.2 (dd, J = 20.7, 31.2 Hz, 1 F), 63.3 (dd, J = 1.9, 32.6 Hz, 1 F). HR-MS (M + H^+) 198.9995, calcd for $C_6H_3F_4O_3$ 190.0018. Anal. Calcd: C, 36.36; H, 1.01. Found: C, 36.29; H, 1.11.

Acknowledgment. We are grateful to the donors of Petroleum Research Fund, administered by the American Chemical Society, and the National Science Foundation (CHE-8822674) for support of this work.

Supplementary Material Available: ^1H NMR spectra for compounds 11, 12, and 2 (3 pages). Ordering information is given on any current masthead page.

Preparation of 1,2-Diketones from Nonenolizable Aliphatic and Aromatic Acyl Chlorides with Diethyl 1-Alkyl(aryl)-1-(trimethylsiloxy)-methanephosphonates1

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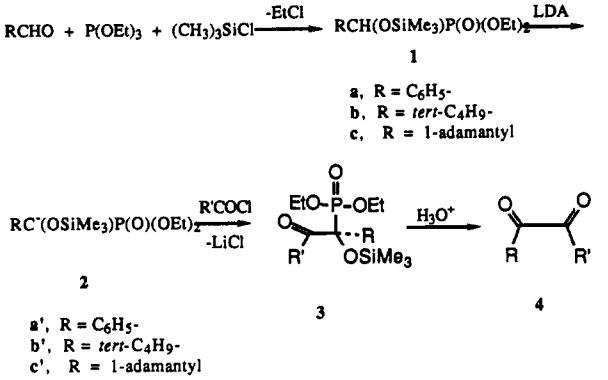
The methods for preparation of 1,2-diketones include acyloin condensation of esters with sodium metal and subsequent oxidation[3] and selenium dioxide oxidation of various monoketones.[3,4] Both methods involve multisteps. α-Diketones are also available through recent oxidative procedures such as the ene reaction of singlet oxygen with alkenes in the presence of titanium alkoxide[5] (followed by epoxide opening) or by the oxidation of acetylenes with $NaIO_4/RuO_2$.[6]

Scheme I



Masked acyl anions have become important synthons in organic synthesis. The most widely used methods for generation of masked acyl anions involve dithio acetals[7] and protected cyanohydrins.[8] More recently[9] the concept of charge affinity inversion[10] has also been applied.

Koenigkramer and Zimmer used the reaction of diethyl 1-phenyl-1-(trimethylsiloxy)methanephosphonate, the precursor of a desired acyl anion, in the presence of lithium diisopropylamide with various ketones to obtain the corresponding α-trimethylsiloxy ketones.[11] We report now a new simple preparation of 1,2-diketones by the reaction of the related acyl anion equivalents, **2**, with acyl chlorides.

The preparation of 1,2-diketones by the reaction of diethyl 1-phenyl(or alkyl)-1-(trimethylsiloxy)methanephosphonates **1** with acyl chlorides is outlined in Scheme I.

Similar to the preparation of diethyl 1-phenyl-1-(trimethylsiloxy)methanephosphonate,[12] diethyl 1-*tert*-butyl-1-(trimethylsiloxy)methane- and 1-(1′-adamantyl)-1-(trimethylsiloxy)methanephosphonates were prepared in nearly quantitative yield (94 and 96%, respectively) from trimethylacetaldehyde and 1-adamantanecarboxaldehyde, respectively, with chlorotrimethylsilane and triethyl phosphite. Subsequent treatment with lithium diisopropylamide (LDA) was carried out at −78 °C to afford the corresponding anions which were then reacted with acyl chlorides to give the desired 1,2-diketones.

Due to the bulk of diethyl 1-*tert*-butyl-1-(trimethylsiloxy)methanephosphonate and diethyl 1-(1′-adamantyl)-1-(trimethylsiloxy)methanephosphonate, the yield of the corresponding 1,2-diketones is subject to steric influence in the reactions with trimethylacetyl chloride and 1-adamantylcarbonyl chloride (Table I). On the other hand, a high yield of benzil was obtained in the reaction of diethyl 1-phenyl-1-(trimethylsiloxy)methanephosphonate with benzoyl chloride which suggests the absence of steric hinderence.

Unlike stronger nucleophiles, diethyl [1-alkyl(aryl)-1-(trimethylsiloxy)methyl]phosphonate anions, **2a–c**, behave as weak nucleophiles which are inert toward esters. Furthermore, **2a–c** also demonstrate unusual inertness toward **3a–c**, presumably due to steric bulkiness. However, the diethyl [1-alkyl-1-(trimethylsiloxy)methyl]phsphonate

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