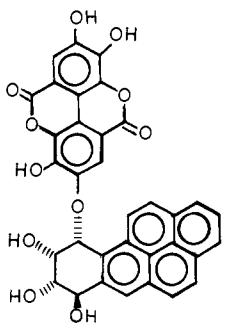
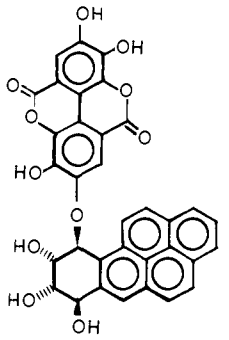
predominant products are a mixture of the cis and trans adducts **3** and **4**.[11] The yield of adducts decreases slightly with decreasing



3



4

pH, possibly because of their partial hydrolysis during the course of reaction at the lower pH values.

Early attempts to characterize these products were complicated by the tendency of the initial product, the unstable cis-adduct **3**, to undergo hydrolysis to tetraols and reaction with excess **1** to yield the more stable trans-adduct **4**. In the absence of added **1**, a mixture of **3** and **4**, consisting predominantly of **3**, was ~50% hydrolyzed to tetraols and **1** after 6 h (pH 7.0, room temperature). In the presence of 10^{-5} M **1**, pH 7.25, the same mixture was converted largely to tetraols and additional trans-adduct **4** after ~7 h. Higher concentrations of **1** accelerated the disappearance of **3** and enhanced formation of **4** relative to tetraols.

To determine the position of substitution on the ellagic acid molecule, we treated adducts **3** and **4** with diazomethane and hydrolyzed the resultant methyl ethers to give trimethyl ethers of ellagic acid. Chromatographic and UV comparison of these esters with synthetic 3,4,4′- and 3,3′,4-tri-*O*-methylellagic acids[12] established that the 3,3′,4-tri-*O*-methyl derivative was produced from both adducts. Hence, the position of substitution is on the 4-hydroxyl of ellagic acid. Adducts involving the 3-position of **1** were not observed.

A change in dioxane concentration from 0.3% to 10% decelerates the reaction of **1** with **2** 12-fold at pH 7.45. This is consistent with an increase in charge separation in the transition state that would be expected for general acid catalysis.[4,13] of epoxide ring opening by a phenolic moiety. Although phenol accelerates the disappearance of **2**,[4a] the second-order rate constant for the reaction of the dianion of **1** (μ 0.1) is >3000 times the analogous rate constant for phenol (μ 0.2). The monoanion and neutral species of **1** are even more reactive and are comparable to hydronium ion. This extraordinary reactivity probably results from specific interactions between the aromatic ring systems[14] of **1** and **2**, although preequilibrium complexation between **1** and **2** in the ground state could not be demonstrated at experimentally practicable concentrations of **1**. In the transition state, a hypothetical complex (Figure 2) having the ellagic acid moiety directly above the aromatic rings of the diol epoxide and on the same side of the molecule as the epoxide oxygen is ideally arranged both for proton transfer to the epoxide oxygen and for cis attack on C_{10} of the benzo[*a*]pyrene moiety.

Ellagic acid is 10 times more potent as an inhibitor of mutagenesis induced by **2** than is riboflavin 5′-phosphate (FMN).[7] Correspondingly, ellagic acid is more effective in bringing about the chemical cleavage of the epoxide ring in **2** than is FMN, which is an extraordinarily effective catalyst for the hydrolysis of **2** to tetraols.[15] The second-order rate constants for the reaction of **2** with the fully protonated and monoanionic forms of **1** are 8-20-fold larger than the corresponding rate constant (204 M^{-1} s^{-1}) for the reaction of **2** with low concentrations of the catalytically active monoanion of FMN. Furthermore, the dianion of FMN is catalytically ineffective, whereas the dianion of **1** still retains two ionizable hydrogens and exhibits a high level of reactivity up to pH 9. Studies are in progess to determine whether ellagic acid can inhibit the tumorigenic activity of **2** in experimental animals.

Registry No. **1**, 58917-67-2; **2**, 476-66-4; **3**, 82933-08-2; **3-AC**, 82933-09-3; **4**, 82977-31-9; **4-Ac**, 82977-32-0.

Supplementary Material Available: Details of the product analyses and the separation and identification of the methylated and acetylated derivatives of **3** and **4** (4 pages). Ordering information is given on any current masthead page.

(10) (a) Yagi, H.; Thakker, D. R.; Hernandez, O.; Koreeda, M.; Jerina, D. M. *J. Am. Chem. Soc.* **1977**, *99*, 1604-1611. (b) Thakker, D. R.; Yagi, H.; Lu, A. Y. H.; Levin, W.; Conney, A. H.; Jerina, D. M. *Proc. Natl. Acad. Sci. U.S.A.* **1976**, *73*, 3381-3385.

(11) Adducts **3** and **4** were purified as their acetates, **3-Ac** and **4-Ac**, and the stereochemistry of these compounds was determined from their NMR spectra. The 100-MHz NMR spectrum of **3-Ac** ($CDCl_3$) is as follows: δ 6.17 (H_8), 5.74 (H_9), 6.80 and 6.84 (H_7 and H_{10}), with $J_{7,8}$ and $J_{9,10}$ = 4-5 and $J_{8,9}$ = 3 Hz; acetoxy protons δ 1.85-2.50 and aromatic protons δ 7.7-8.4. The assignments for the methine protons H_7-H_{10} are consistent with the corresponding chemical shifts and coupling constants for the acetate of the cis adduct of phenol and **2** (ref 10a). Satisfactory resolution of H_8 and H_9 of **4-Ac** could not be obtained even at 500 MHz in $CDCl_3$ but was obtained in acetone-d_6: δ 6.92 (H_7), 5.92 (H_8), 6.04 (H_9), 7.04 (H_{10}), with $J_{7,8}$ = 9, $J_{8,9}$ = 2, and $J_{9,10}$ = 3 Hz; acetoxy protons δ 2.0-2.35 and aromatic protons δ 7.95-8.30 and 8.6. The large value of $J_{7,8}$ for this compound requires that the acetoxy groups at C_7 and C_8 be diequatorial. Thus the axial ellagic acid substituent at C_{10} must be cis to the acetoxy group at C_7 and hence must have resulted from attack on the face of **2** that is opposite to the epoxide oxygen.

(12) The synthetic trimethylellagic acids were prepared by reaction of diazomethane with 4,4′ and 3,3′-di-*O*-methylellagic acids (Jurd, L. *J. Am. Chem. Soc.* **1959**, *81*, 4606-4610) followed by chromatographic purification (see Supplementary Material for details). Ultraviolet spectra observed by us for 3,3′,4-tri-*O*-methylellagic acid and its anion (in 1:9 dioxane-water) agree with previously reported spectra in ethanol (Jurd, L.; Palmer, K. J.; Stitt, F.; Shoolery, J. N. *J. Am. Chem. Soc.* **1959**, *81*, 4620-4623).

(13) The general acid-catalyzed hydrolyses of **2** by the monanions of phosphate and guanosine 5′-monophosphate are subject to similarly large solvent effects: see ref 4d.

(14) Enhanced stabilization of the cation-like transition state for epoxide cleavage by a π-π donor-acceptor interaction with the electron-rich ellagic acid molecule is an attractive possibility. Molecules capable of acting as π-electron donors have been shown to accelerate the acetolysis of 2,4,7-trinitro-9-fluorenyl-*p*-toluenesulfonate via complex formation: Colter, A. K.; Wang, S. S.; Megerle, G. H.; Ossip, P. S. *J. Am. Chem. Soc.* **1964**, *86*, 3106-3113. Colter, A. K.; Hui, S. H. *J. Org. Chem.* **1968**, *33*, 1935-1940.

(15) Wood, A. W.; Sayer, J. M.; Newmark, H. L.; Yagi, H.; Michaud, D. P.; Jerina, D. M.; Conney, A. H. *Proc. Natl. Acad. Sci. U.S.A.* **1982**, *79*, 5122-5126.

Homolytic Carbocyclization by Use of Heterogeneous Supported Organotin Catalyst. A New Synthetic Route to 2-Alkoxytetrahydrofurans and γ-Butyrolactones

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Received May 12, 1982

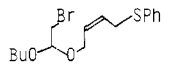
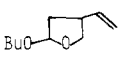
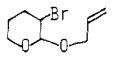
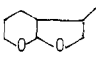
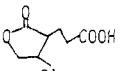
The synthesis or functionalization of γ-butyrolactones has been currently studied as a key synthetic reaction.[1] All such lactones have been prepared by polar reactions but not by radical ones.[2] Although radical reactions seem to be attractive in some aspects, their synthetic applications, especially to the C-C bond formations, are very restricted presumably due to the serious side reactions characteristic to them. For example, radical cyclization of 5-hexen-1-yl radical or its related species such as 3-oxa-5-hexen-1-yl radical has been investigated well, but only from the mechanistical viewpoint.[3]

(1) Wolfe, J. F.; Ogliaruso, M. A., "The Chemistry of Acid Derivatives"; Patai, S., Ed.; Wiley: New York, 1979; Part 2, Chapter 19.

(2) The only exceptional example is a free radical cyclization of *N*-iodoamides to lactones. Barton, D. H. R.; Beckwith, A. L. J., Goosen, A. *J. Chem. Soc.* **1965**, 181.

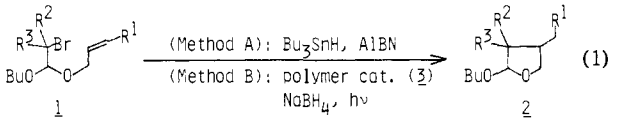
0002-7863/82/1504-5564$01.25/0

Table I

	1			2(%)		4 (%)
	R^1	R^2	R^3	Method A	Method B	
a	H	H	H	59	91	87
b	Me	H	H	59	86	79
c	n-Pr	H	H	65	96	91
d	H	$-(CH_2)_5-$		50	73	70
e				 78	91	 95
f				 68	84	 81

In connection with our continuous study on the synthetic applications of the homolytic processes,[4] we report here a new method for the preparation of γ-butyrolactones via 2-alkoxytetrahydrofurans, in which the key step consists of the homolytic carbocyclization.

We found that bromoacetals (**1**)[5] cyclized to five-membered cyclic acetals (**2**) but not to six-membered ones under homolytic conditions. To a solution of **1** and a catalytic amount of azobisisobutyronitrile (AIBN) (1 mol %) in dry benzene was added dropwise an equivalent of tributyltin hydride at room temperature or in some cases at 50 °C in ~30 min (eq 1; 0.25 M concentration

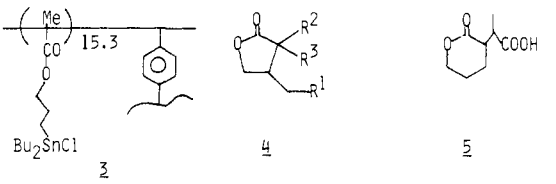


(1)

of Bu_3SnH). After stirring for 2 h, distillation of the mixture gave the desired **2** (Method A, Table I).

The moderate yields may be largely attributable to the purification procedure to remove the somewhat troublesome tributyltin bromide as a byproduct.[6] This led us to use a polymer-supported organotin,[7] since insoluble polymeric supports were claimed to enable easy separation of synthetic products by simple filtration.[8] Thus, the catalytic reaction using cross-linked polymer catalyst (**3**)[9] (0.1 equiv) in the presence of sodium tetrahydroborate ($NaBH_4$)[10] (1.5 equiv) in a degassed benzene–ethanol mixed solvent afforded **2** in improved high yield under UV irradiation (100-W high-pressure mercury lamp in a Pyrex tube) at room temperature for 30 min. Removal of the polymer catalyst **3** by filtration followed by the usual workup afforded the sufficiently pure **2** free from organotin species without fractional distillation (Method B, Table I).[11] Another advantage is emphasized by the successful reuse of the recovered catalyst **3** for several times without loss of its activity.[12]

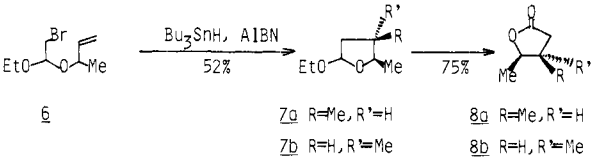
A one-pot procedure for the conversion of 2-alkoxytetrahydrofurans into γ-butyrolactones has been reported by use of *m*-chloroperbenzoic acid/boron trifluoride etherate.[13] Our choice is a Jones reagent,[14] which enables the easy separation of butoxy fragment by its conversion to butanoic acid. Thus, Jones reagent (2.2 equiv) was added slowly to an ice-cooled solution of **2** in acetone. After the complete addition, excess isopropyl alcohol was added. Usual workup followed by distillation gave **4** (Table I).



The present cyclization method seems to be particularly useful for the C–C bond formation at the sterically bulky carbon, since such bond formation in polar reactions, involving carbocations or carbanions and often accompanying the elimination reaction or skeletal rearrangements, are well-known to be strongly affected by the steric hindrance and generally gave poor results. This was visualized in the successful preparation of spirocyclic γ-butyrolactone (**4d**).

The method, when coupled with the intramolecular S_H' process,[16] produced the cyclic acetal (**2e**), which gave β-vinyl-γ-butyrolactone **4e**. Bicyclic acetal **1f** obtained from dihydropyran yielded exclusively γ-butyrolactone **4f** in 81% yield, but not six-membered lactone **5**.[17]

The highly stereoselective homolytic cyclization was observed in the following case. Thus, cis-lactone **8a** (eq 2) was predominantly obtained (96:4 **8a**/**8b**). This ratio was estimated by the known ^1H NMR (CCl_4) and GPC,[18] i.e., *o*-methine protons appeared at δ 4.08 and 4.56 in **8a** and **8b**, respectively.



(2)

The present method provides an attractive entry to the preparation of 2-alkoxytetrahydrofurans and γ-butyrolactones from easily available allylic alcohols and vinyl ethers with various

(3) Beckwith, A. L. J.; Blair, I.; Phillipou, G. *J. Am. Chem. Soc.* **1974**, *96*, 1613. Smith T. W.; Butler, G. B. *J. Org. Chem.* **1978**, 43, 6. Walling, C.; Cioffari, A. *J. Am. Chem. Soc.* **1972**, *94*, 6059.

(4) Ueno, Y.; Aoki, S.; Okawara, M. *J. Am. Chem. Soc.* **1979**, *101*, 5415.

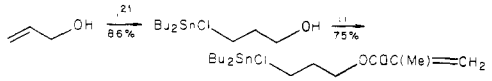
(5) **1** and **6** were prepared in 75–92% yield by the reaction of an equimolar amount of *N*-bromosuccinimide with vinyl ether in the presence of excess of allylic alcohol at -20 to -50 °C for 2 h. **1e** was prepared in 92% yield by using excess of vinyl ether as solvent.

(6) Several devices have been reported to achieve the effective separation of organotin species from synthetic products. These methods, however, were not always sufficient in our case. Saigo, K.; Morikawa, A.; Mukaiyama, T. *Bull Chem. Soc. Jpn.* **1976**, *49*, 1656. Berge, J. M.; Roberts S. M. *Synthesis* **1979**, 471.

(7) Two reports on the polymeric organotin hydride have appeared. Weinshenker, N. M.; Crosby, G. A.; Wong, J. Y. *J. Org. Chem.* **1975**, *40*, 1966. Schumann, H.; Pachaly, B. *Angew. Chem., Int. Ed. Engl.* **1981**, *20*, 1043.

(8) For a review, see: Mathur, N. K.; Narang, C. K.; Williams, R. E. "Polymers as Aids in Organic Chemistry"; Academic Press: New York, 1980.

(9) The methacrylate monomer was prepared as follows:



(i) Bu_2SnH_2/Bu_2SnCl_2, (ii) $ClCOC(Me)=CH_2$/4-dimethylaminopyridine

The monomer was copolymerized with styrene in the presence of 5% divinylbenzene and a catalytic amount of AIBN in benzene at 70 °C for 113–143 h in a sealed tube. The content of tin moiety was estimated by the elemental analysis.

(10) The catalytic system (Bu_3SnCl–$NaBH_4$) was employed for the reduction of organic halides (RX → RH). Corey, E. J.; Suggs, J. W. *J. Org. Chem.* **1975**, *40*, 2554.

(11) The catalytic reaction using Bu_3SnCl in place of polymer **3** also required fractional distillation to remove the organotin species after the reaction, which caused a decreased yeild of **2**.

(12) Polymer catalyst was recovered after quenching with methyl iodide. The soluble copolymer **3'** without cross-linking with divinylbenzene gave poor results for reuse. The deactivation may be attributable to the coupling of tin radical leading to distannane. "Site-isolation"[22] in the cross-linked polymer **3** greatly prevents it from such coupling.

(13) Grieco, P. A.; Oguri, T.; Yokoyama, Y. *Tetrahedron Lett.* **1978**, 419.

(14) Craig, J. C.; Horning, E. C. *J. Org. Chem.* **1960**, *25*, 2098.

(15) All new compounds had satisfactory spectral and analytical data.

(16) Ueno, Y.; Chino, K.; Okawara, M. *Tetrahedron Lett.* **1982**, *23*, 2575.

(17) **4f**: IR 1760 cm^{-1} (C=O); ^{13}C NMR ($CDCl_3$) δ 31.357 (CH_2COOH).

(18) Kanetsuna, H.; Nonaka, T.; *Denki Kagaku* **1979**, *47*, 422.

substitution patterns. Furthermore, the method represents a clear-cut example of the synthetically useful C–C bond formation via a homolytic process. It also demonstrates the obvious advantage of the polymer supported reagent over the corresponding low molecular weight one in the aspects of the product yield and reuse of the reagent, as well as the well-claimed simplicity of the procedure. C–C bond formations using polymer catalyst that has been hitherto reported have been very few.[19] The stereoselective homolytic carbocyclization may provide a new synthetic methodology. Studies on this line and the application to the natural product synthesis are now in progress.[20]

Registry No. 1a, 82918-70-5; **1b**, 82918-71-6; **1c**, 82918-72-7; **1d**, 82932-66-9; **1e**, 82918-73-8; **1f**, 73746-50-6; **2a**, 82918-74-9; **2b**, 82918-75-0; **2c**, 82918-76-1; **2d**, 82918-77-2; **2e**, 82918-78-3; **2f**, 82918-79-4; **3**, 82918-69-2; **4a**, 1679-49-8; **4b**, 16496-51-8; **4c**, 22530-99-0; **4d**, 82918-80-7; **4e**, 43142-60-5; **4f**, 82918-81-8; **6**, 82918-82-9; **7a**, 82918-83-0; **8a**, 10150-95-5; **8b**, 10150-96-6; Bu_3SnH, 688-73-3; $NaBH_4$, 16940-66-2.

(19) Trost, B. M.; Keinan, E. *J. Am. Chem. Soc.* **1978**, *100*, 7779.
(20) Our result suggests the high possibility of the assymetric synthesis of γ-butyrolactones having two assymetric centers at β and γ positions starting from a chiral alcohol [CH_2=CHCH*(Me)OH].
(21) Neumann, W. P.; Pedain, J. *Tetrahedron Lett.* **1964**, 2461.
(22) Crowley, J. L.; Rapoport, H. *Acc. Chem. Res.* **1976**, *9*, 135–144.

Tetrathiazyl Tetrakis(*F-tert*-butoxide)

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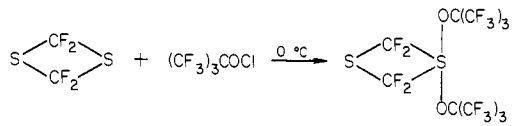
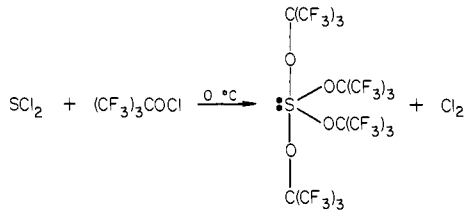
Received May 20, 1982

Extending our examination of the reactions of *F-tert*-butyl hypochlorite, we now report our studies with non-carbon-containing sulfur systems such as tetrasulfur tetranitride. Earlier we had demonstrated that $(CF_3)_3COCl$ can be caused to react with sulfur compounds in several ways,[2] e.g.

(a) oxidative addition



(b) oxidative displacement and oxidative addition



(c) oxidative displacement

$$CF_3S(O)Cl + (CF_3)_3COCl \xrightarrow{0\ ^\circ C} (CF_3)_3COS(O)CF_3 + Cl_2$$

The reactions of S_4N_4 are unusually diverse and continue to hold considerable fascination for the synthetic chemist. Tetrasulfur tetranitride appears to be susceptible to attack by free radicals to form S-tetrasubstituted or -disubstituted derivatives, e.g., with $(CF_3)_2NO \rightarrow N_4S_4(ON(CF_3)_2)_4$,[3,4] with $S_2O_6F_2 \rightarrow N_4S_4(OSO_2F)_2$,[5] where the latter contains the tetrathiazyl cation, $S_4N_4^{2+}$, or with $Cl_2 \rightarrow N_4S_4Cl_2$.[6]

Tetrasulfur tetranitride and *F-tert*-butyl hypochlorite react at 0 °C to form a white solid identified as $N_4S_4(OC(CF_3)_3)_4$ in 98% yield. After 8 or 9 h at 0 °C, the bright orange S_4N_4 is completely decolorized, and gaseous chlorine, which is recovered quantitatively, is visible in the reaction flask.

$$S_4N_4 + 4(CF_3)_3COCl \xrightarrow{0\ ^\circ C} N_4S_4(OC(CF_3)_3)_4 + 2\ Cl_2$$

If the reaction is carried out at 25 °C, small amounts of CF_3Cl and $(CF_3)_2CO$, which result from the decomposition of the hypochlorite, are also found in the vapor over the solid. The product was found to be insoluble or very slightly soluble in water, diethyl ether, carbon tetrachloride, and methylene chloride. It is soluble in CCl_3F and CCl_2FCF_2Cl and may be recrystallized from either one. In some runs a small amount of a yellow substance was formed upon contact with air.

Sublimation occurs at 70 °C at a pressure of 50 μm. In a sealed capillary, the sublimed solid melts at 161–162 °C accompanied by slight discoloration. The ^{19}F NMR spectrum contains a singlet at ϕ −70.7. In the mass spectrum, a parent ion is not observed, but an ion that may be assigned to $(M - OC(CF_3)_3)^+$ at m/e 889 is recorded. Other appropriate fragments are observed such as $S_4N_4^+$, $S_3N_3^+$, $S_2N_2^+$, and SN^+. This supports the retention of the eight-membered ring.

The properties of the three known $N_4S_4(R_f)_4$ (R_f = F, $(CF_3)_2NO$, $(CF_3)_3CO$) compounds are remarkably similar, e.g., $(M - R_f)^+$ is the largest m/e, and they are white crystals that sublime in vacuo and melt with decomposition or discoloration and are essentialy insoluble in all common organic solvents. While $N_4S_4(ON(CF_3)_2)_4$ does not appear to be even wetted by water, both $N_4S_4F_4$ and $N_4S_4(OC(CF_3)_3)_4$ are slowly attacked by water. The structure of the new tetrakis(*F-tert*-butoxide) is expected to be essentially identical with that of $N_4S_4(R_f)_4$ (R_f = F, $(CF_3)_2NO$) with the $(CF_3)_3CO$ groups bonded to the sulfur atoms. The S_4N_4 ring should be opened, slightly destroying any transannular interactions but retaining bond lengths associated with three coordinated sulfur and two coordinated nitrogen.

Thus, $(CF_3)_3COCl$ can be used as an excellent oxidative addition reagent for introducing *F-tert*-butoxy groups into cyclic lower valent sulfur compounds under sufficiently mild conditions to prevent ring opening.

Experimental Section. Gases were manipulated in a conventional Pyrex vacuum apparatus equipped with a Heise Bourdon tube gauge and a Televac thermocouple gauge. Infrared spectra were obtained by using a 5 cm stainless steel cell with KBr windows or as solids between KBr discs on a Perkin-Elmer 599 spectrometer. ^{19}F NMR spectra were recorded with a JEOL FX90Q spectrometer operating at 84.26 MHz. Chemical shifts are relative to CCl_3F. Mass spectra were measured with a Hitachi Perkin-Elmer RMU-6E mass spectrometer at 25 eV. Elemental analyses were performed at the University of Idaho or by Beller Laboratories, Göttingen, West Germary.

F-tert-Butyl hypochlorite was synthesized by the literature method.[8] S_4N_4 was freshly recrystallized from benzene.

To S_4N_4 (0.105 g, 0.571 mmol) in a 50-mL Pyrex flask equipped with a Kontes Teflon stopcock and Teflon-coated stirring bar was added $(CF_3)_3COCl$ (1.053 g, 3.90 mmol). The mixture was stirred at 0 °C for 8–9 h. After the volatle materials were removed under vacuum, the white solid, $N_4S_4(OC(CF_3)_3)_4$, weighed 0.632 g (0.562 mmol) for a 98% yield. Chlorine (0.081 g; theoretical 0.081 g) was the only other product obtained. The infrared spectrum of $N_4S_4(OC(CF_3)_3)_4$ taken between KBr discs had bands at 1305 (sh), 1274 (br vs), 1250 (s), 1234 (sh), 1198 (m), 1188 (m), 1060 (vs), 998 (s), 979 (s), 778 (m), 754 (w), 745

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