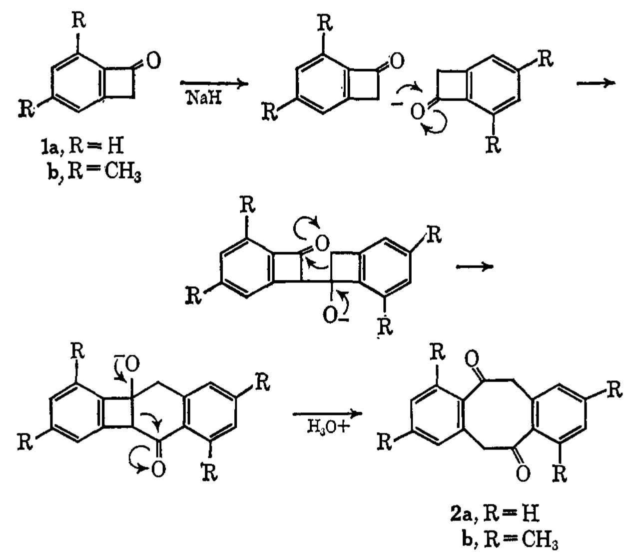
Scheme I



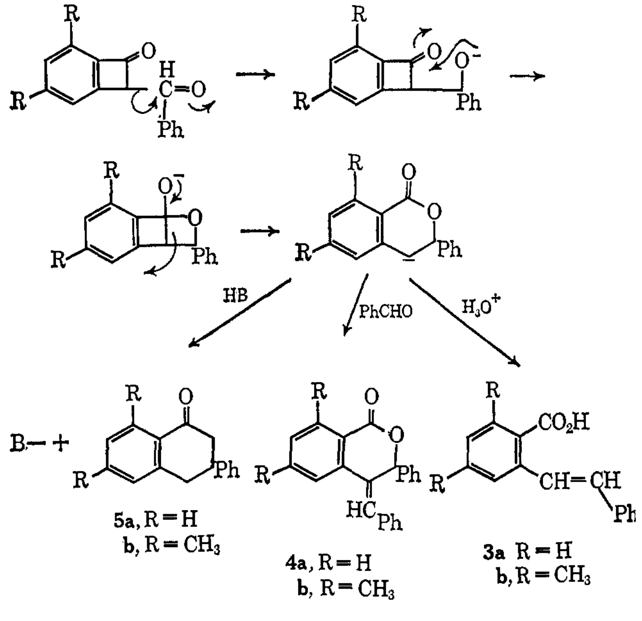
1a, R = H
b, R = CH₃

2a, R = H
b, R = CH₃

products (~50%) carboxylic acids **3a** and **3b**, along with trace amounts of four lactones, **4a**,**4b** and **5a**,**5b**. The mass spectral molecular weights of **3a** and **3b** (**3a**, *m*/*e* 224 ($C_{15}H_{12}O_2$); **3b**, *m*/*e* 252 ($C_{17}H_{16}O_2$)) and nmr spectra (**3a**, τ −2.3 broad s, 2.08 and 3.14, AB quartet, J = 16.5 cps, and 2.06–2.94 m; relative intensities 1:2:9; **3b**, τ 0.6 broad s, 2.90 doublet of an AB quartet, J = 16.5 cps, 2.4–3.06 m, 7.75 s, and 7.84 s; relative intensities 1:1:8:3:3) are consistent with the stilbene-2-carboxylic acid and 3,5-dimethylstilbene-2-carboxylic acid[5] structures indicated. Conclusive proof of structure **3a** could be obtained by comparison of the physical properties of this compound to those reported[8] for stilbene-2-carboxylic acid.

The mass spectral molecular weights of **4a**[5] and **4b**[5] (**4a**, *m*/*e* 312 ($C_{22}H_{16}O_2$); **4b**, *m*/*e* 340 ($C_{24}H_{20}O_2$)), infrared spectra (**4a**, ν 1726 (C=O), 1273 and 1225 (C—O) cm^{-1}; **4b**, ν 1720 (C=O), 1242 and 1236 (C—O), and nmr spectra (**4a**, τ 2.1–2.8 m and 3.37 s; relative intensities 15:1; **4b**, τ 2.3–2.8 m, 3.0 s, 3.4 s, 7.42 s, and 7.6 s; relative intensities 12:1:1:3:3) are consistent with the structures indicated. The two remaining lactones, **5a** and **5b**, were not isolated as crystalline compounds, but the infrared spectra (**5a**, ν 1726 (C=O), 1273, 1225 (C—O) cm^{-1}; **5b**, ν 1720 (C=O), 1242 and 1236 (C—O) cm^{-1}) and nmr spectra (**5a**, τ 1.8–3.0 m, 4.37 (X part of an ABX), and 6.75 (AB part; $\Delta\nu_{AB}$ = 24 cps, J_{AB} = 17 cps, J_{AX} = 12.5 cps, and J_{BX} = 2.5 cps); relative intensities 9:1:2; **5b**, τ 2.2–2.8 m, 4.64 (X part of an ABX), 6.97 (AB part; $\Delta\nu_{AB}$ = 24 cps, J_{AB} = 17 cps, J_{AX} = 12.5 cps, and J_{BX} = 2.5 cps), 7.35 s, and 7.73 s; relative intensities 7:1:2:3:3) are consistent with the structures assigned. The products arising from the reaction of benzocyclobutenone and benzaldehyde in the presence of NaH can be accounted for according to Scheme II.[7]

Scheme II



5a, R = H
b, R = CH₃

4a, R = H
b, R = CH₃

3a R = H
b, R = CH₃

Addition of **2b** to a suspension of NaH in DMF containing either methyl iodide or methyl chloroformate yielded only alkylated or acylated dimer. Therefore the carbonyl of benzocyclobutenone is more reactive toward condensation than a typical ketone, but less reactive than an aldehyde. Although these data do not establish the enolate character of the anion of benzocyclobutenone, work presently in progress to obtain quantative data concerning the acidity of benzocyclobutenone as well as additional base-catalyzed reactions of this system may resolve this point.

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[8] D. F. DeTar and L. A. Carpino, *J. Am. Chem. Soc.*, 78, 475 (1956).

Ylides from Salts of Sulfoximines[1,2]

Sir:

Sulfur ylides are now well established as reagents for organic synthesis. In large part the synthetic achievements with these reagents have originated from the work of Corey and coworkers.[3] This preliminary report describes a new series of oxosulfonium salts and some reactions of their derived ylides.

A series of N,N-dimethylaminoalkylaryloxosulfonium fluoroborates[4] has been prepared by the sequence shown in Scheme I. The oxosulfonium salts

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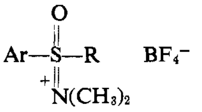
[1] (a) Part X in the series Chemistry of Sulfoxides and Related Compounds; (b) Part IX: C. R. Johnson, J. C. Sharp, and W. G. Phillips, *Tetrahedron Letters*, 5299 (1967).

[2] We gratefully acknowledge support by the National Science Foundation (Grant No. GP-5944).

[3] (a) E. J. Corey and M. Chaykovsky, *J. Am. Chem. Soc.*, 87, 1353 (1965); (b) E. J. Corey, M. Jautelat, and W. Oppolzer, *Tetrahedron Letters*, 2325 (1967); (c) E. J. Corey and M. Jautelat, *J. Am. Chem. Soc.*, 89, 3912 (1967).

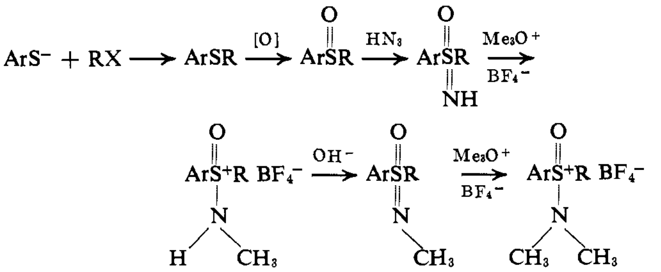
[4] Systematic nomenclature for the nitrogen analogs of sulfoxides and sulfones is not well established. The name sulfone imine has been proposed (R. G. Laughlin and W. Yellin, *ibid.*, 89, 2435 (1967)) for compounds previously known as "sulfoximines"; accordingly, the final product shown in Scheme I would be termed N,N-dimethylamino alkyl aryl sulfone iminium fluoroborate. The origin of the latter name is more obvious from the resonance form.



$$\mathrm{Ar{-}\overset{\overset{\displaystyle O}{\|}}{\underset{\underset{\displaystyle N(CH_3)_2}{\overset{+}{\|}}}{S}}{-}R}\quad \mathrm{BF_4^-}$$

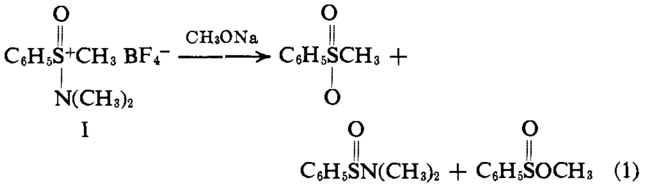
From the work in our laboratories on such salts it appears that the oxosulfonium nomenclature is more descriptive.

Scheme I

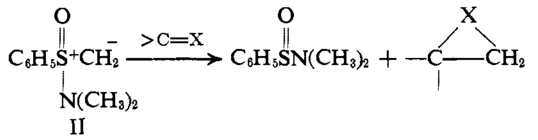


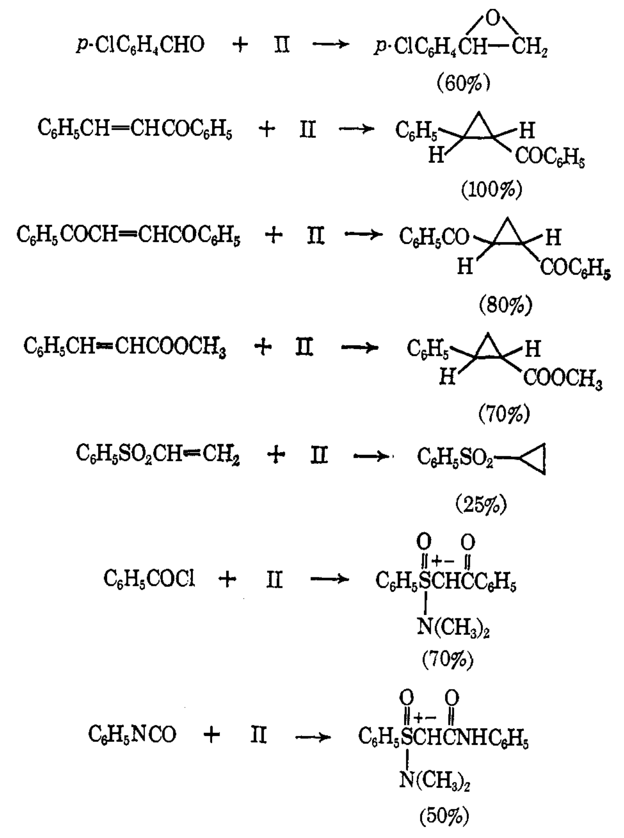
and the N-alkylsulfoximines are new classes of compounds.[5]

The reaction of N,N-dimethylaminomethylphenyloxosulfonium fluoroborate (I), mp 118–119°, δ (CH_2Cl_2) 3.1 (singlet, $N(CH_3)_2$), 3.95 (singlet, SCH_3), 7.5–8.2 (C_6H_5), with sodium methoxide in refluxing methanol is illustrated in eq 1. Presumably the methyl phenyl sulfone is formed in a reaction involving direct attack of the methoxide on the sulfonium sulfur, the sulfinamide by attack on the S-methyl or by decomposition of the methylide, and the ester from the initially formed amide.

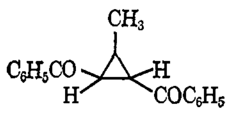


Treatment of I at room temperature with sodium hydride in dimethyl sulfoxide resulted in the rapid and quantitative evolution of hydrogen with the formation of a slightly yellow solution of ylide II. Addition of water to a solution of the ylide in dimethyl sulfoxide or tetrahydrofuran results in a strongly basic solution from which the original salt (I) could be regenerated by neutralization with fluoroboric acid. Illustrative examples of the reactions of ylide II with carbonyl compounds and electrophilic olefins are shown below.[6] In most cases the highly polar sulfinamide could be removed by passing a benzene solution of the reaction products through a short column of silica gel.

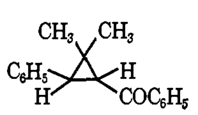




A number of similar syntheses have also been achieved employing ylides derived from ethyl and isopropyl salts; *e.g.*



(50%)



(60%)

This sulfur ylide chemistry is particularly noteworthy in that extensive structural variation can be easily achieved. This has not been readily possible with other sulfur ylides, although Corey[3b,c] has recently had notable successes with the sulfonium ylides derived from diphenylsulfonium alkylides.[7] It appears that the ylides derived from our salts may be considerably more stable and may be generated under milder conditions than those employed for the simpler sulfonium ylides. An intriguing facet of this chemistry lies in the fact that optically active salts and ylides should be readily accessible. Such reagents might well yield optically active oxiranes and cyclopropanes. Extensive investigations of the chemistry of the new types of compound reported here and related compounds are under way in our laboratories.

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(5) Salts of the general structures $[R_2S(O)NH_2]^+X$ and $[R_2S(O)N{=}PR_3]^+X$ have been reported (R. Appel, H. Fehlaber, D. Hanssgen, and R. Schollhorn, *Chem. Ber.*, **99**, 3108 (1966); T. W. Rave and T. J. Logan, *J. Org. Chem.*, **32**, 1629 (1967)). In a subsequent paper we will report on the preparation of additional examples of N-alkylated sulfoximines and certain N-alkyl derivatives of sulfilimines.

(6) The reactions illustrated are similar to those previously reported for dimethyloxosulfonium methylide. For a summary of sulfur ylide chemistry see A. W. Johnson, "Ylide Chemistry," Academic Press Inc., New York, N. Y., 1966. See also H. Metzger and H. Konig, *Z. Naturforsch.*, **18b**, 987 (1963); W. E. Truce and V. V. Badiger, *J. Org. Chem.*, **29**, 3277 (1964); S. R. Landor and N. Punja, *J. Chem. Soc., C*, 2495 (1967).

(7) For additional examples of the reactions of diphenylsulfonium alkylides see A. W. Johnson, V. J. Hruby, and J. L. Williams, *J. Am. Chem. Soc.*, **86**, 9181 (1964).

(8) Alfred P. Sloan Research Fellow, 1965–1968.

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The Total Synthesis of Some Monomeric Vinca Alkaloids: *dl*-Vincadine, *dl*-Vincaminoreine, *dl*-Vincaminorine, *dl*-Vincadifformine, *dl*-Minovine, and *dl*-Vincaminoridine[1]

Sir:

The transannular cyclization reaction developed in these laboratories has provided a general synthetic

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(1) Presented at a Symposium on the Chemistry of Natural Products, University of West Indies, Kingston, Jamaica, Jan 2–5, 1968.