Tetrabutylammonium Phenyl(phenylsulfonyl)methylide: A Chiral Metal-free "Carbanion"

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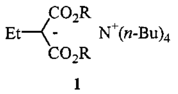
The treatment of benzylphenylsulfone with tetrabutylammonium hydroxide results in the formation of the title compound tetrabutylammonium phenyl(phenylsulfonyl)methylide. An X-ray structural analysis of the racemate shows the presence of a chiral metal-free carbanion in the form of an intimate ion pair. Stabilization of the conformational enantiomer occurs by a three-point interaction with two of the α-methylene moieties of the tetrabutylammonium ion, CH···O hydrogen bonding being the driving force. The ion pair is capable of initiating the anionic polymerization of acrylates.

The term "carbanion" has traditionally been used by organic chemists to denote carbon-based anionic species such as Grignard reagents, alkyllithium compounds or lithium enolates, as well as other deprotonated CH-acidic compounds such as the α-lithio forms of nitriles, isonitriles, nitroalkanes, sulfoxides or phosphonates.[1] Detailed structural studies of organolithium compounds have shown that true carbanions are not present.[2] Instead, lithium is closely bonded to the anionic part of the reagents, the compounds often occurring in solution and in the solid state as dimers, tetramers or higher aggregates. In contrast, metal-free systems such as tetralkylammonium salts of such CH-acidic compounds as ketones, esters, nitriles or nitro-alkanes were traditionally believed to be "naked" carbanions,[3] occurring as intermediates in phase-transfer catalyzed alkylations, Michael additions[4] and various other reactions.[5] Their exceptional reactivity in the metal-free polymerization of acrylates and (meth)acrylates also seemed to indicate special electronic properties.[6] However, systematic X-ray structural studies of the tetrabutylammonium salts of various esters, ketones, lactones, nitriles, and nitroalkanes clearly revealed that "naked" carbanions are not involved.[7] Rather, the anions undergo strong H-bonding with the α-methylene groups of the tetrabutylammonium cations as is the case for compounds **1−6**, the species generally existing as dimers or higher aggregates in solution and in the solid state. Structural studies of dimers such as **1** show that supramolecular ion pairs are involved.[7a]



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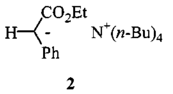

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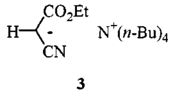

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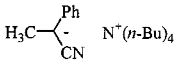

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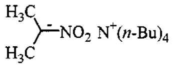

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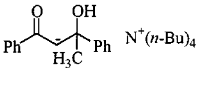

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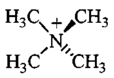
In all of these compounds the negatively charged ions interact with positively charged ions[7] by [CH···O] or [CH···N] H-bonding, which are common structural motifs in other classes of crystalline compounds.[8] The reason why tetraalkylammonium ions are such good H-donors can be attributed to favorable positive charge delocalization. Thus, ab initio calculations have shown that the positive charge in the parent tetramethylammonium species is not localized on nitrogen, as traditionally assumed by organic chemists (cf. **7a**),[9] but delocalized evenly over the four methyl groups (cf. **7b**).[10] Indeed, the hydrogen atoms also bear a significant portion of the positive charge, whereas nitrogen is calculated to be neutral. As a result the methyl groups are expected to be more acidic than those in trimethylamine $(CH_3)_3N$.



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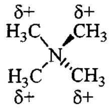

7b

In this paper we describe the synthesis, crystal structure and aggregation state of the α-sulfonyl carbanion **8**. Included is a brief report on the behavior of this compound as an initiator in the metal-free polymerization of acrylates. α-Sulfonyl carbanions have a long history as useful synthetic intermediates in organic synthesis.[11] Boche has reported the crystal structure of several α-lithio-sulfones,[2a] including that of α-(phenylsulfonyl)benzyllithium-TMEDA,[12] which is the lithio-analog of **8**. The only metal-free mono-α-sulfonyl carbanion previously characterized by X-ray crystallography is compound **9**, first reported by Gais.[13]

The synthesis of compound **8** was readily accomplished by treating the CH-acidic sulfone **10** with commercially available $Bu_4N^+(OH)^-$ and removing the water azeotropically. Recrystallization from DMF afforded yellow

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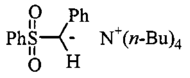
crystals of the racemate suitable for an X-ray structure analysis.



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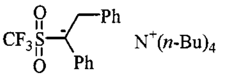

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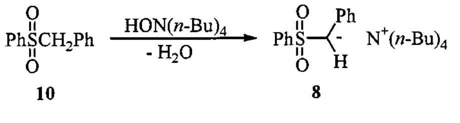

Several features of the solid state structure of **8** (Figure 1) deserve comment. As in the case of the tetrabutylammonium enolates **1**, **2**, **3** and **6**, and the nitronate **5**, the anion and cation interact through [CH···O] hydrogen bonding.[7] In contrast to the α-sulfonyl carbanion **9**, however, which crystallizes with the tetrabutylammonium cation to form layers of parallel, extended polymers,[13] compund **8** forms an intimate ion pair with one tetrabutylammonium cation in such a way that both oxygen atoms of the sulfonyl group interact with H-atoms of two α-methylene units [O1···C5 3.390(4), O2···C1 3.553(4) Å].[14] In addition, the relatively short interionic C17···C1 distance of 3.696(4) Å indicates that a three-point interaction is involved, similar to that observed in the crystal structure of the tetrabutylammonium enolate of phenyl(2-hydroxy-2-phenyl)propyl ketone (**6**), where a C(H)···C$^-$ distance of 3.557(2) Å is found.[7e]

A cryoscopic study in benzene showed that the compound is also monomeric in solution ($\mathrm{MW}_{\mathrm{calcd}}$ = 474; $\mathrm{MW}_{\mathrm{exp}}$ = 485). With the exception of potassium bis(trimethylsilyl)methyl(phenyl)sulfone-[18-crown-6][15] and lithium bis(methyl)-methyl(phenyl)sulfone-[2.1.1]cryptand,[16] all other structurally studied α-sulfonyl carbanions are dimers or higher aggregates. In the case of the lithium compound the carbanion is free and unassociated whereas in the potassium compound there is a two-point interaction between the oxygen atoms of the sulfone and the metal.

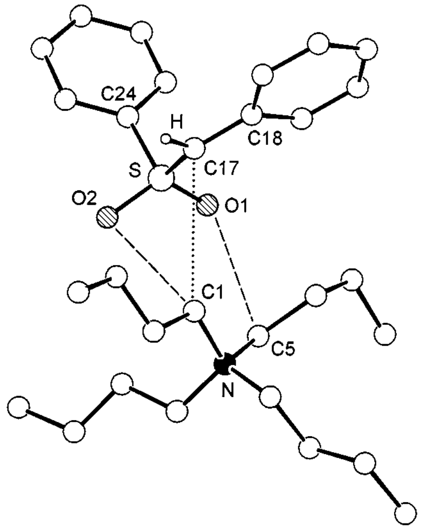

Figure 1. Molecular structure of **8**, showing three-point interaction between the chiral α-sulfone carbanion and the α-methylene groups on one face of the tetra-*n*-butylammonium cation [O1···C5 3.390(4), O2···C1 3.553(4), C1···C17 3.696(4) Å]

The most important structural details of **8** are listed in Table 1. As a consequence of the stabilizing interaction between the lone electron pair of the anionic C-atom and the sulfonyl moiety, the C−S bond length [1.667(3) Å] is significantly shortened relative to those in neutral sulfones (typically, 1.786 Å).[17] At the same time the S−O bond lengths [mean, 1.442(3) Å] are longer than in neutral sulfones (typically, 1.436 Å). The benzylic H-atom attached to C17 was located in the X-ray experiment and its position well refined [C−H 0.92(3) Å, U_{H} = 0.08 Å^2]. Although the H-atom could not be located with the same degree of accuracy as the heavier atoms, its position clearly shows that the carbanion adopts a *gauche* arrangement with respect to the sulfonyl group. Figure 2 shows a Newman projection along the C17−S bond.

As can be seen in Figure 2, the carbanion center is pyramidal and the sum of the absolute values of the torsion angles C24−S−C17−H [+78(2)°] and C24−S−C17−C18 [−76(1)°] is significantly less than 180°, in contrast to other phenyl-substituted sulfonyl carbanions which usually have

Table 1. Selected interatomic distances and angles in **8**[a]

Bond lengths (Å)		Angles (°)	
S−O1	1.442(2)	O1−S−O2	117.7(2)
S−O2	1.442(3)	O1−S−C17	112.0(2)
S−C17	1.667(3)	O1−S−C24	104.5(2)
S−C24	1.771(3)	O2−S−C17	108.6(2)
C17−C18	1.427(4)	O2−S−C24	104.7(2)
C17−H	0.92(3)	C17−S−C24	108.8(2)
		S−C17−C18	127.4(3)
		S−C17−H	115(2)
		C18−C17−H	113(2)
		C1−N−C5	108.4(2)
		Torsion angles (°)	
		C24−S−C17−C18[b]	−76(1)
		C24−S−C17−H[c]	78(2)
		O1−S−C17−C18	39(1)
		O1−S−C17−H	−167(2)
		O2−S−C17−C18	171(1)
		O2−S−C17−H	−35(2)
Interionic distances (Å)			
O1···C5	3.390(4)		
O2···C1	3.553(4)		
C1···C17	3.696(4)		

[a] In the crystal both enantiomers are found. − [b] Defined as β in ref.[2a] and α_2 in ref.[19] − [c] Defined as α_1 in ref.[19]

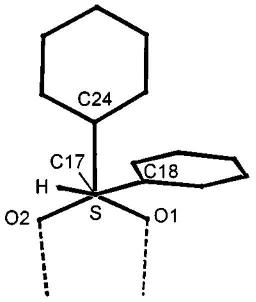

Figure 2. Newman projection of **8** along the C17(carbanion center)−S bond; dashed lines indicate the interionic [CH···O] hydrogen bonding interactions; torsion angles are given in Table 1

Table 2. R−S−C^-−R_x torsion angles in structurally characterized mono-sulfonyl carbanions, R−SO_2−C^-(R_1)(R_2)

R	R_1	R_2	R−S−C^-−R_1 α (°)	R−S−C^-−R_2 β (°)	Cation	Compound[a]	Reference
Ph	H	H	−81(3)	81(3)	Li^+	CSD51495[b]	18
Ph	$SiMe_3$	$SiMe_3$	−116.0	69.6	K^+	CSD52970[b]	15
Ph	Me	Me	−71.7(4)	75.8(4)	Li^+	SOCYIS	16
Ph	CH = CH_2	H	−76(1)	78(3)	Li^+	FAGFOI	19
Ph	Me	Me	−72.9(4)	66.3(4)	Li^+	GAVZAE	20
Ph	Ph	H	−70.4(6)	calcd	Li^+	DIBXIV	12
Ph	Ph	Me	−79.8	89.4	Li^+	GAVYUX	20
CF_3	Ph	CH_2Ph	−84.0	93.1	Li^+	SEMHEX	13
CF_3	Ph	CH_2Ph	−80.3/−83.8[c]	96.7/92.6[c]	Li^+	VAVKAE	21
Ph	Ph	H	−76(1)	78(2)	NBu_4^+	**8**	This work
CF_3	Ph	CH_2Ph	−77.1	95.2	NBu_4^+	**9** (SEMHIB)	13

[a] REFCODE in the Cambridge Crystallographic Database. − [b] Fachinformationszentrum Energie, Physik, Mathematik GmbH, D-7514 Eggenstein-Leopoldshafen 2, Germany. − [c] Crystallographically independent ions.

an almost planar carbanion center. Table 2 gives comparable angles for the structurally characterized monosulfone carbanions {R−SO_2−C^-(R_1)(R_2)} (planar: $|\alpha| + |\beta| = 180°$). More significantly, the larger lobe of the lone pair is situated *antiperiplanar* to the substituent R on the S-atom, i.e., between the O-atoms in the Newman projection, and points in the direction of the proton on C1, the α-methylene C atom of the tetrabutylammonium cation that forms a [CH···O] hydrogen bond with O2.

It thus appears that the sulfonyl carbanion in **8** is an example of a conformational enantiomer that is stabilized by a three-point interaction with one face of a tetrabutylammonium cation. Of course, the compound is racemic. Here, as in almost all structurally characterized lithiosulfones, the carbanion lone pair is *antiperiplanar* to the substituent on the sulfonyl group, consistent with the observation that α-chiral sulfones undergo base catalyzed H/D-exchange with retention of configuration.[22]

The suitability of the sulfonyl stabilized anion **8** as an initiator for the anionic polymerization of *n*-butylacrylate (BA) and methylmethacrylate (MMA) at room temperature was investigated according to procedures described earlier.[6] In the case of BA an exothermic reaction occurred immediately on addition of monomer solution, but yields were generally poor [typically, M_n(expt) = 1500 for all M_n(calcd), D = 1.05, yield = 50%]. Quantitative conversions were also not obtained with the less reactive MMA. Here the yields were higher, at least for the lower target molecular weights [typically, M_n(expt) = 2500; M_n(calcd) = 3600, D = 1.47, Yield = 80%], and there was no observable induction time or associated rise in temperature, indicating a relatively slow initiation. These observations contrast strongly with those obtained using other initiators, such as metal-free malonates.[6,7f] This is somewhat surprising in view of the higher basicity of the phenyl(phenylsulfonyl)methylide carbanion relative to these compounds and may be due to the particular stabilizing interaction of the tetrabutylammonium cation.

In conclusion, we have shown that the phenyl(phenylsulfonyl)methylide carbanion **8** is stabilized asymmetrically by a three-point interaction with one face of a tetrabutylammonium cation. Molecular weight studies indicate that the resulting chiral ion-pair is also stable in solution. The formation of a stabilized intimate ion pair may be the reason why the metal-free α-sulfonyl carbanion **8**, in spite of its higher basicity, is a poorer initiator for the polymerization of methyl methacrylate than metal-free malonates of type **1**.

Experimental Section

All operations were performed in flame-dried reaction vessels under an argon atmosphere using standard Schlenk techniques. *n*-Butyl acrylate (BA) and methyl methacrylate (MMA) were purified by distillation over calcium dihydride under high vacuum just before use. Toluene and THF were refluxed over sodium tetraethylaluminium under inert gas conditions (argon). Gel permeation chromatographic (GPC) measurements were performed at 20°C with THF as eluent, a flow rate of 1.0 mL min^{-1}, and with a differential refractor detector and a variable-wavelength UV detector. The molecular weight calibration curve was obtained with PBA and PMMA as standards. ^1H and ^{13}C NMR spectra were measured in [D_6]-DMSO on a Bruker AM-200 spectrometer, where the chemical shifts were determined with respect to the solvent as internal standard. Cryoscopic measurements were performed by Dornis & Kolbe (Mülheim/Ruhr, Germany) in benzene using a Beckmann thermometer. Quantitative GC measurements were made on quenched polymer solutions.

Tetrabutylammonium phenyl(phenylsulfonyl)methylide (8): Tetrabutylammonium hydroxide (20 mmol, 20 mL, 1 M in methanol; Aldrich) was added to a 250 mL flask equipped with two 100 mL dropping flasks and connected via a liquid N_2 trap to a membrane vacuum pump. The solution was diluted with 50 mL toluene and subsequently 6 × 20 mL of toluene were added and each time the volume reduced under a vacuum of 15 bar, taking care to ensure that the volume did not go below 70 mL. After the addition of a final 50 mL volume of toluene, phenylbenzylsulfone (**10**) (4.646 g, 20 mmol) dissolved in 40 mL THF was added dropwise to the tetrabutylammonium hydroxide solution over 2 h. At the same time the solvent was continuously distilled into the trap. During the procedure the volume of the solution was not allowed to go below 100 mL. After addition of all the phenylbenzylsulfone solution, the mixture was reduced to 20 mL and dried in vacuo overnight, yield 35% (3.32 g). − ^{1}H NMR (200 MHz, [D$_6$]-DMSO): δ = 0.92 (t, ^3J = 7.0 Hz, 12 H, CH$_3$), 1.29 (sextet, ^3J = 7.2 Hz, 8 H, N−CH$_2$−CH$_2$−C*H*$_2$), 1.55 (m, 8 H, N−CH$_2$−C*H*$_2$), 3.15 (m, 8 H, N−CH$_2$), 6.09 (s, 1 H, −CH), 6.68 (s, 2 H, *p*-CH), 7.06-7.36 (m, 4 H, *m*-CH), 7.49-7.76 (m, 4 H, *o*-CH). − ^{13}C NMR (50 MHz, [D$_6$]-DMSO): δ = 13.4 (CH$_3$), 19.1 (N−NCH$_2$−CH$_2$−*C*H$_2$), 23.0 (N−CH$_2$−*C*H$_2$), 57.5 (N−CH$_2$), 64.3 (−C), 112.2 (*p*-C benzyl), 117.2, 124.1, 127.3, 127.8, 129.0, 130.8, 133.7, 138.3, 144.6, 154.3 (C-aromatics).

Crystal Structure Analysis of *rac*-8:[23] $[C_{13}H_{11}O_2S]^-[C_{16}H_{36}N]^+$, M_r = 473.74, yellow prism, crystallized from dimethylformamide, crystal size 0.18 × 0.49 × 0.49 mm, a = 9.008(1), b = 19.681(2), c = 16.501(1) Å, β = 102.810(7)°, U = 2852.6(5) Å^3, T = 293 K, monoclinic, space group $P2_1/n$ (No. 14), Z = 4, D_c = 1.10 g cm^{-3}, μ = 0.14 mm^{-1}. CAD4 diffractometer, $Mo\text{-}K_\alpha$ X-radiation, λ = 0.71069 Å. 5133 measured reflections, no absorption correction, 4917 unique, 2597 (gt) with $I > 2.0\sigma(F_o^2)$. The structure was solved by direct methods (SHELXS-97) and refined by full-matrix least-squares (SHELXL-97) on F^2 for all data with Chebyshev weights to R = 0.057 (gt), wR = 0.164 (all data), 302 parameters, S = 1.01, H atom attached to C17 isotropic [U_H = 0.08(1) Å^2], remaining H atoms riding, max shift/error 0.001, residual $\Delta\rho_{max}$ = 0.346 e Å^{-3}.

Polymerizations: As reported previously for similar polymerizations,[7] the initiator **8** was placed in a flame-dried flask under an argon atmosphere and dissolved in carefully dried THF. Monomer was dissolved in the same weight of solvent and added dropwise at room temperature from a syringe pump (1 mL min^{-1}) into the stirred solution of the initiator. The polymerization was quenched 1 h after the monomer addition by injection of 1 mL of 1 M HCl. Poly(*n*-butyl acrylates) were diluted with diethyl ether and washed with water in a separating funnel. The organic layers were dried over MgSO$_4$, filtered and the solvents were evaporated under vacuum. Poly(methyl methacrylates) were slowly poured into water/methanol (4:1). The freshly precipitated polymers were filtered and dried under vacuum in a drying oven at 45 °C.

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[23] The crystallographic data (without structure factors) of **8** have been deposited as supplementary publication CCDC-124968 at the Cambridge Crystallographic Data Centre. Copies of the data can be obtained from: The Director, CCDC, 12 Union Road, Cambridge CB2 1EZ, UK. (Fax: + 44-1223/336-033; E-mail: deposit@ccdc.cam.ac.uk).

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