Unexpected Rearrangements Induced by the Metal Cation in the Reduction of Cycloocta[*b*]naphthalene with Alkali Metals in Ethers. An EPR and ENDOR Study

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A set of structurally different radicals has been detected by EPR in the reduction of cycloocta[*b*]naphthalene (**1**) by potassium in 1,2-dimethoxyethane. The results differ from those previously reported for the reduction in hexamethylphosphoric triamide, where the radical anion $\mathbf{1}^{\bullet -}$ yields the dianion $\mathbf{1}^{2-}$ by further reaction with potassium. In a second reduction step, an apparently symmetry-forbidden ring closure leads to benzo[*b*]biphenylene (**2**), shown by EPR spectrum of $\mathbf{2}^{\bullet -}$. By subsequent reduction, a bond fragmentation takes place in the cyclobutadiene ring, leading to the 2-phenylnaphthalene radical anion $\mathbf{3}^{\bullet -}$. Finally, the EPR spectrum of the radical anion of an unknown compound **4** is observed, the structure of which is tentatively proposed to be a dipotassium salt of dibenzoheptafulvene. It was possible to show that $\mathbf{4}^{\bullet -}$ arises from $\mathbf{2}^{\bullet -}$ by a different opening process of the cyclobutadiene ring, followed by rearrangement. This study was extended to other alkali metals (lithium and sodium), and to other ethereal solvents (2-methyltetrahydrofuran and tetrahydrofuran). Strong ion association determines the course of the reduction. Mechanisms for the successive reactions observed are proposed.

Cyclic hydrocarbons, as well as ring-fused conjugated systems containing $4n\pi$ electrons, may acquire aromaticity either by oxidation to the corresponding dications or by reduction to the dianions. Cyclooctatetraene and related compounds are good examples of such $4n\pi$ systems.

The parent cyclooctatetraene dianion and some substituted derivatives have been extensively studied. The tub-shaped ground-state geometry for the cyclooctatetraene ring has been unambigously established by X-ray crystallographic structure determination of cyclooctatetraenecarboxylic acid.[1] By electrolytic reduction, or with alkali metals, these compounds yield first the planar or nearly planar radical anions,[2–4] and then the planar dianions. As expected, the X-ray crystallographic structure determination of the potassium salt of 1,3,5,7-tetramethylcyclooctatetraene dianion in diglyme, $\{K[(CH_3OCH_2CH_2)_2O]\}_2[C_8H_4(CH_3)_4]$, shows the eight membered ring in a planar conformation as an almost regular octagon with an average C–C bond length of 1.407 Å.[5] ^{1}H NMR in tetrahydrofuran (THF) confirms this by displaying a single peak.[6]

There is also indirect evidence for the planarity of the cyclooctatetraene ring in the radical anion of dibenzo[*a*,*e*]cyclooctene, and even more in its dianion. In the electroreduction of dibenzo[*a*,*e*]cyclooctene, as studied by cyclic voltammetry,[7] the first reduction step (corresponding to the conversion of the tub-shaped parent to the presumably, already planar radical anion) was found to be slower than the second step leading to the planar dianion. Moreover, although a persistent radical anion is obtained[8] by reduction of dibenzo[*a*,*e*]cyclooctene with lithium, sodium or potassium in THF at room temperature, the same radical anion, when obtained by reduction with potassium in dimethoxyethane (DME) in the temperature range from 193 K to 223 K, has been reported to decay with increasing temperature. The decay is accompanied by the appearance of a new and unidentified paramagnetic species.[9] In the reduction with potassium in 2-methyltetrahydrofuran (MTHF) this radical has not been observed.[9] The ^{1}H NMR spectra of the corresponding dianions in solution show the protons at the cyclooctatetraene ring, shifted to lower field than the corresponding proton peaks in the neutral compound.[8] This behaviour is characteristic of an induced ring current in the cyclooctatetraene ring, as occurs in aromatic molecules, and suggests strong electron delocalization over the ring. EPR and thermodynamic studies for the reduction of the related compounds benzocyclooctene[10] and cycloocta[*b*]naphthalene,[10] with potassium in hexamethylphosphoric triamide (HMPA), also point to the planarity of the corresponding radical anions and dianions.

Except for studies carried out in highly dissociating solvents such as HMPA or dimethylformamide (DMF), which favour free radical anions and dianions, the studies reported in less polar solvents such as THF or DME often show evidence of a poorly understood ion-pairing effect. We have been greatly interested in such ion-pairing effects on reactivity. In this paper we report new results obtained in the reduction of cycloocta[*b*]naphthalene (**1**) with alkali metals in ethereal solvents, at various temperatures.

Results

When cycloocta[*b*]naphthalene (**1**) is reduced with lithium, sodium or potassium in DME over the temperature range 183–213 K, persistent paramagnetic solutions are obtained with well resolved EPR spectra, corresponding to the radical anion $\mathbf{1}^{\bullet -}$. When the sample is allowed to warm to room temperature, a fast decrease of the EPR intensity is observed. The observed EPR spectrum is identical with the one reported earlier for the reduction of **1** with potassium in HMPA at room temperature.[10]

The results of further reduction of $\mathbf{1}^{\bullet -}$ in DME depend on the alkali metal used. With sodium or potassium very different EPR spectra are observed, corresponding to a structurally different radical anion. This EPR spectrum is identical with the one reported for the radical anion of benzo[*b*]biphenylene $\mathbf{2}^{\bullet -}$.[11] In contrast, when $\mathbf{1}^{\bullet -}$ reacts with lithium, the primary radical gradually disappears and finally a diamagnetic solution is obtained.

Subsequent reduction with more potassium leads to another paramagnetic species without the C_{2v} symmetry of $\mathbf{1}^{\bullet -}$ and $\mathbf{2}^{\bullet -}$.

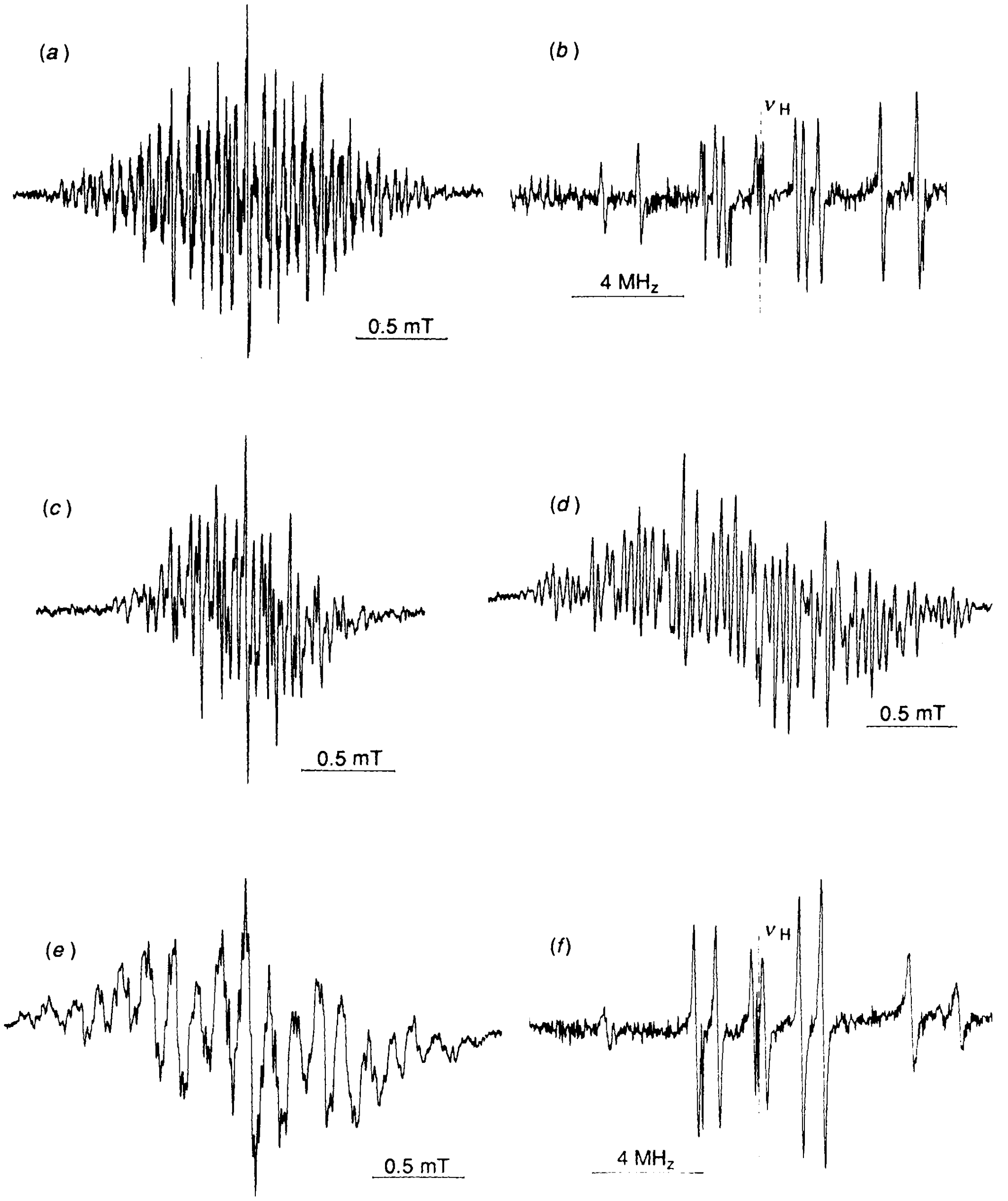


Fig. 1 Experimental EPR and ENDOR spectra in DME at 193 K: (*a*) $\mathbf{1}^{\bullet -}K^+$ (EPR); (*b*) $\mathbf{1}^{\bullet -}K^+$ (ENDOR); (*c*) $\mathbf{2}^{\bullet -}K^+$ (EPR); (*d*) $\mathbf{3}^{\bullet -}K^+$ (EPR); (*e*) $\mathbf{4}^{\bullet -}K^+$ (EPR); and (*f*) $\mathbf{4}^{\bullet -}K^+$ (ENDOR)

The EPR spectrum is identical with that reported for the 2-phenylnaphthalene radical anion $\mathbf{3}^{\bullet -}$.[12] After further contact of the solution with the alkali-metal mirror, the EPR signal corresponding to $\mathbf{3}^{\bullet -}$ disappears and a new EPR spectrum is obtained, the hyperfine pattern of which denotes a higher symmetry. For the sake of simplicity, from now on, this species will be referred as $\mathbf{4}^{\bullet -}$.

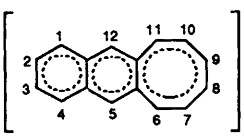
If **1** is reduced with potassium in the comparatively less polar solvents, THF or MTHF in the same temperature range, it becomes experimentally difficult to record the EPR spectra of the different paramagnetic species previously described for the reduction in DME. Nevertheless the different colours of all the paramagnetic species described, namely $\mathbf{1}^{\bullet -}$–$\mathbf{3}^{\bullet -}$, can still be seen and the corresponding EPR spectra can be followed in rapid scan mode, which means that the same structural changes are occurring, but at a faster rate than in DME. The only persistent paramagnetic species giving a well resolved EPR spectrum is $\mathbf{4}^{\bullet -}$. This behaviour is also observed for reduction of **1** with potassium in DME at room temperature.

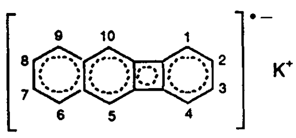
In order to substantiate the proposed structures, independent reduction studies of **2** and **3** were carried out. When **2** was reduced by potassium in DME, the primary radical anion was $\mathbf{2}^{\bullet -}$ (identical with that obtained from **1** in the second step) and the radical anion $\mathbf{3}^{\bullet -}$ was obtained by further reduction. Finally, after subsequent reduction, $\mathbf{3}^{\bullet -}$ disappears and $\mathbf{4}^{\bullet -}$ is obtained. In the reduction of **3**, using the same experimental conditions, only the primary radical anion $\mathbf{3}^{\bullet -}$ was observed. Further reduction leads to a diamagnetic solution. This experiment rules out the possibility that $\mathbf{3}^{\bullet -}$ is a precursor of $\mathbf{4}^{\bullet -}$.

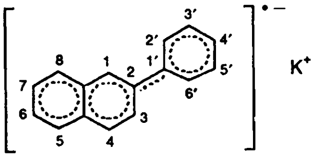
We have also investigated the possibility of $\mathbf{3}^{\bullet -}$ being generated from $\mathbf{2}^{\bullet -}$ by increasing the temperature, but without further reduction. In fact, with increasing temperature the EPR signal of $\mathbf{2}^{\bullet -}$ disappears, but only a very weak signal of $\mathbf{3}^{\bullet -}$ begins to appear. Its intensity increases abruptly by further contact with the alkali metal.

The EPR spectra of the different radical anions are shown in Fig. 1 and the corresponding hyperfine splittings (hfs) are in Table 1. The assignments of hfs are based on McLachlan's spin-density calculations. The EPR studies have been further substantiated by ENDOR and triple-resonance studies, that allowed the hfs for $\mathbf{1}^{\bullet -}$ to $\mathbf{4}^{\bullet -}$ to be determined with more confidence. In Fig. 1 the ENDOR spectrum of $\mathbf{1}^{\bullet -}$ is included as

Table 1 Experimental hfs (mT) $1^{\bullet -}$ to $4^{\bullet -}$ obtained by reduction of **1** with alkali metals in DME at 193 K. Figures in parentheses denote molecular positions







$1^{\bullet -}M^{+\,a,b}$	$2^{\bullet -}K^{+\,c}$	$3^{\bullet -}K^{+\,d}$	$4^{\bullet -}K^{+\,e,f}$
−0.410 (6,11)	0.247 (2,3)	0.544 (1)	−0.522 (2H)
−0.315 (8,9)	0.152 (7,8)	0.382 (8)	−0.399 (2H)
−0.152 (7,10)	0.141 (5,10)	0.352 (4)	0.167 (2H)
−0.116 (5,12)	0.110 (1,4)	0.296 (5,6)	0.110 (2H)
−0.094 (1,4)	0.047 (6,9)	0.231 (4′)	0.015 (2H)
−0.008 (2,3)		0.159 (7)	
		0.134 (6′)	
		0.063 (2′)	
		0.043 (3)	
		0.032 (3′)	
		0.013 (5′)	

[a] M = Li, Na, K. [b] The signs were determined by ENDOR/TRIPLE resonance. The assignments are based on theoretical spin-density calculations. [c] The hfs measured are identical with those referred to in ref. 11 and the assignments are those indicated in this reference. [d] As *c* from ref. 12. [e] The multiplicities were determined by ENDOR/TRIPLE resonance. For the structure of **4**, see discussion in the text. [f] $g = 2.0040 \pm 0.0001$.

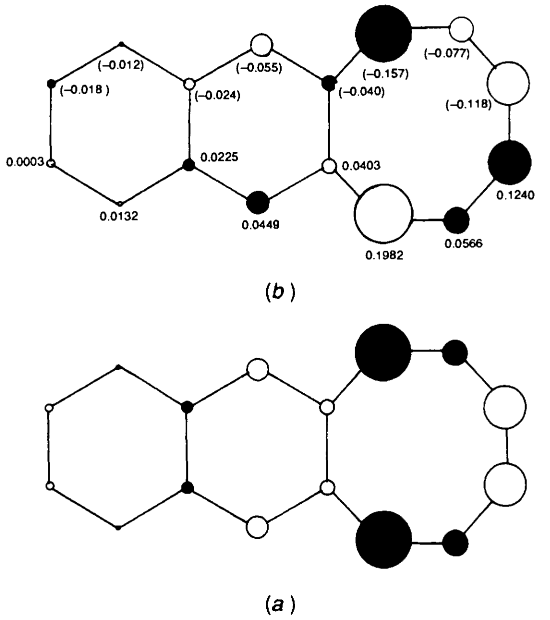


Fig. 2 (*a*) HOMO of **1** and (*b*) SOMO (semi-occupied MO) of $1^{\bullet -}$. The areas of the circles are proportional to the squares of the linear combination of atomic orbital coefficients. Filled and blank circles symbolize the different signs of these coefficients. Numbers within parentheses represent excess charge densities and the others represent spin densities.

well as the ENDOR spectrum from the last paramagnetic species described as $4^{\bullet -}$. For the sake of brevity, the ENDOR spectra of the remaining paramagnetic species have been omitted. The ENDOR spectrum of $3^{\bullet -}$ has been reported previously.[12] The relative signs of all the hfs indicated in Table 1 were determined by studying the enhancement of ENDOR signals in general triple-resonance experiments.

Discussion

In the reduction of **1**, with the different alkali metals, in DME, THF or MTHF all the EPR spectra recorded at 193 K refer to solvent-separated ion pairs or even free radical anions. The observed enhanced kinetic stability at lower temperatures derives from the full delocalization of the negative charge over the planar molecule. In fact the hyperfine pattern was shown to be independent of the nature of the cation. As the temperature increases, the distance between the ions in each ion pair tends to shorten. This effect increases from lithium to the less solvated sodium and further to potassium. The proximity of a positive charge affects the negative-charge distribution over the delocalized anion. In contact ion pairs the negative charge thus becomes more localized at the carbon atoms which are nearest to the cation. Particularly for $1^{\bullet -}K^{+}$, where the observed rate of decay of the EPR signal is the greatest, the ion pair is expected to be the tightest and thus the negative charge to be more localized. The cyclooctatetraene ring therefore becomes less planar and the radical anion more prone to undergo the subsequent structural changes leading to the experimentally observed rearranged radicals.

It is not clear whether this structural change takes place in the radical anion, a diamagnetic carbanion, or the dianion. In any case, the great influence of the interaction with the alkali counter-ion, has to be considered as a determining factor. Indeed, in those reductions carried out in more polar solvents and/or by means of alkali metals with smaller atomic radii, where free anions may result, the subsequent reduction of $1^{\bullet -}$ leads to the long-lived 1^{2-} and no rearranged radicals, as in the course of the reduction by potassium in DME.

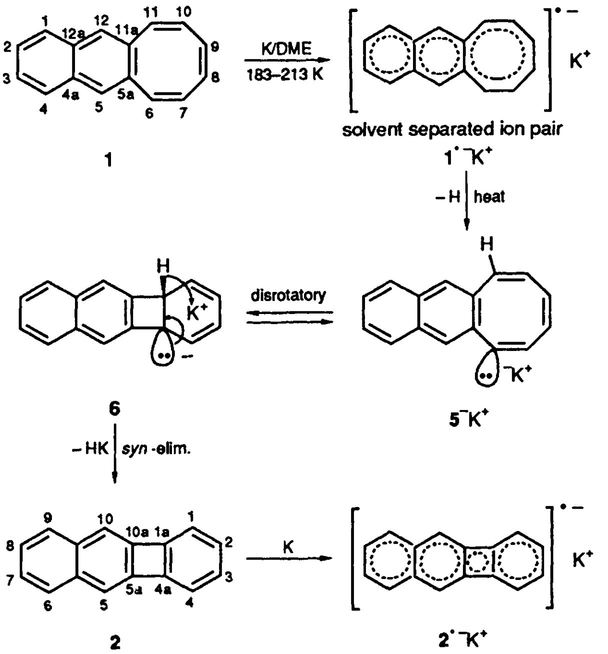
The fact that the ion pair reacts faster than the free ion strongly suggests that the transition state must involve a high charge concentration. This high negative charge could be stabilized by the close proximity of the counter-ion. Although we have no direct evidence, a possible mechanism can be envisaged for the conversion $1^{\bullet -}$ into $2^{\bullet -}$, by means of potassium in DME. Assuming the electrocyclic ring closure to be a concerted reaction, the orbital symmetry must be conserved. In the radical anion the frontier orbital has the symmetry shown in Fig. 2(*b*) where the charge and spin distributions are also indicated. The more reactive positions will be the C-6 and C-11 atoms, *i.e.* in the cyclooctatetraene ring, where the highest charge and spin densities lie. It is evident that the six-electron disrotatory reaction is a symmetry-forbidden process in the radical anion. The same applies to the dianions, since one is dealing with the same frontier orbital [Fig. 2(*b*)].

Nevertheless some change must have occurred. If one looks

Table 2 π Bond orders from HMO calculations

	Bond order	
	$2^{\bullet-}$	2^{2-}
C(4a)–C(4b)	0.360	0.491
C(10a)–C(10b)	0.360	0.491
C(4b)–C(10a)	0.411	0.294
C(10b)–C(4a)	0.449	0.303
Other C–C bonds	⩾0.466	⩾0.410

for possible analogy, the most obvious is the reduction of fluorene with potassium in THF. It is known that the fluorene radical anion obtained at 183 K decomposes at 223 K to yield the fluorenide carbanion,[13] which is subsequently reduced to the respective radical dianion. Similarly we may consider that, by loss of a hydrogen atom to the medium, $1^{\bullet-}K^+$ is converted into 5^-K^+ where potassium (which is taking the place of the hydrogen atom) is now more σ-bonded, and the cyclooctatetraene ring more tub-shaped. It is from either C-6 or C-11 (which have the highest charge and spin density) that this hydrogen abstraction is most probable. An immediate internal electron reorganization (intramolecular π–σ electron transfer) may occur, leading to the carbanion 5^-. As the electron pair now remains in a σ orbital, the relevant π-molecular orbital (π-MO) for the carbanion 5^- has the symmetry of the ground-state highest occupied molecular orbital (HOMO) that is represented in Fig. 2(*a*). The six-electron disrotatory ring closure shown in Scheme 1 is now symmetry-allowed. After the cyclization a *syn*-elimination of H^-K^+ in the ion pair can occur, to yield **2** (ElcB mechanism). Further reduction leads to $2^{\bullet-}$. This whole sequence is represented in Scheme 1.



Scheme 1

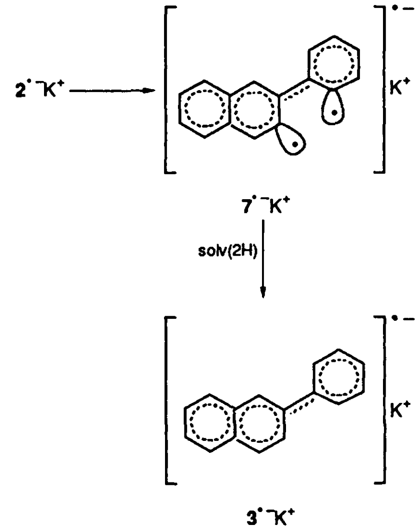
The last paramagnetic species observed $3^{\bullet-}$ and $4^{\bullet-}$ may originate either from the radical anion $2^{\bullet-}M^+$ or from the dianion $2^{2-}M_2^+$. The most labile bonds in both cases lie in the four-membered ring, where the charge densities have the highest values (as well as the highest spin densities in the radical anion).[11] While $3^{\bullet-}$ must result from scission of the C(4a)–C(4b) or C(10a)–C(10b) bond, the formation of $4^{\bullet-}$ must involve the breaking of either the C(10b)–C(4a) or the C(4b)–C(10a) bond. Otherwise it would be more difficult to explain the higher symmetry of $4^{\bullet-}$ shown by its EPR spectrum (five pairs of equivalent protons). Although $3^{\bullet-}$ was generated earlier, the observed symmetry of $4^{\bullet-}$ rules out the possibility of **4** coming directly from **3**. A faster rate of formation of **3** relative to the reaction leading to **4** (due to any kinetic effect) may be sufficient to prevent the simultaneous observation of $3^{\bullet-}$ and $4^{\bullet-}$.

Even when the dianion is not formed in detectable concentrations, there may be tiny quantities present in equilibrium with the radical anion [eqn. (1)]. At the higher temperatures,

$$2(2^{\bullet-}M^+) \rightleftharpoons 2 + 2^{2-}M_2^+ \qquad (1)$$

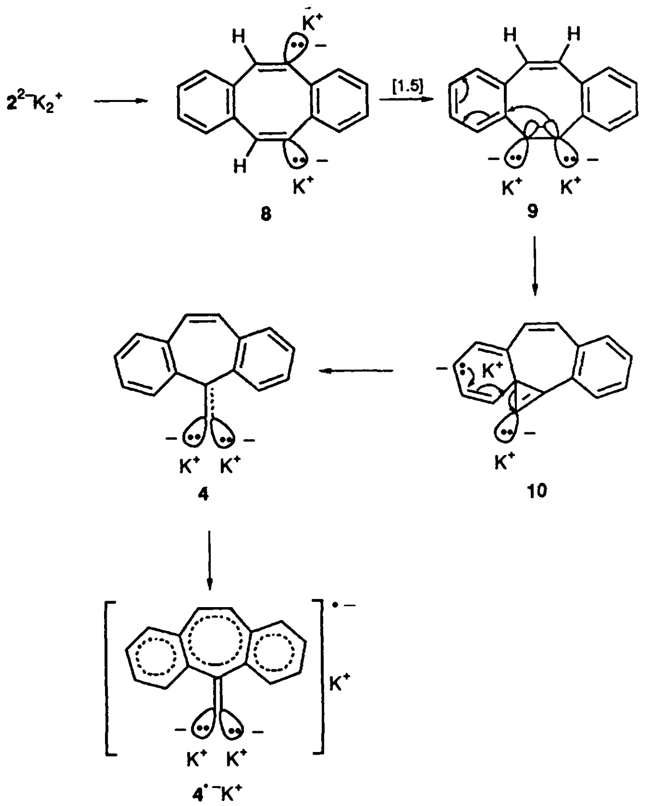
the dianion concentration should increase as a result of less ion solvation in the dianion and consequent increased entropy.

In order to discuss the preferential direction of cyclobutadiene ring opening in $2^{\bullet-}$ or 2^{2-}, bond order calculations by using Hückel molecular orbitals (HMO), have been made. In Table 2 the calculated C–C bond orders for all the bonds in the cyclobutadiene ring for the radical anion and for the dianion are shown. The calculated bond orders point to a faster C(4a)–C(4b) bond cleavage in the radical anion (lowest bond order = 0.360) and a faster C(4b)–C(10a) or C(10b)–C(4a) bond cleavage in the dianion (bond orders of 0.294 and 0.303, respectively). Therefore one is led to consider the radical ion pair $2^{\bullet-}K^+$ as the precursor of $3^{\bullet-}$ and dianionic $2^{2-}K_2^+$ as the precursor of $4^{\bullet-}$, without excluding completely the hypothesis of $2^{\bullet-}K^+$ being a precursor of $4^{\bullet-}$. The faster generation of $3^{\bullet-}K^+$ *via* the C(4a)–C(4b) bond scission in $2^{\bullet-}$ (shown in Scheme 2) is not only supported by the above-mentioned bond-order calculations which point to the C(4a)–C(4b) bond scission, but also by the possibility of a rapid stabilization of the resulting radical $7^{\bullet-}K^+$ by abstraction of two hydrogen atoms from the medium, yielding immediately $3^{\bullet-}K^+$. On the other hand, if **4** results from single bond fragmentation [C(4b)–C(10a) or C(10b)–C(4a)] in $2^{\bullet-}K^+$, followed by hydrogen abstraction, **4** would have to be either dibenzo[*a*,*e*]cyclooctene, or its dihydroderivative, or the initial product cycloocta[*b*]naphthalene, or its dihydroderivative. Neither of these structures correspond to the observed hyperfine pattern for $4^{\bullet-}$ (five pairs of equivalent hydrogens). It thus appears that the initial bond cleavage yielding **8** is followed by subsequent rearrangement and intramolecular hydrogen shift (Scheme 3). A [1,5] hydrogen shift may have a pericyclic mechanism favoured by the fact that the eight-membered ring may adopt a tub-shaped conformation.



Scheme 2

A transition state where the hydrogen is in contact with both



Scheme 3

ends of the chain at the same time favours the thermally allowed suprafacial [1,5] migration yielding **9**. A [1,2] hydrogen shift, on the contrary, seems to be a more unusual process either in radicals or in carbanions. In the present case it is probable that an ion-pair or diradical mechanism may be operative, with the metal playing a determinative role. When the metal ion is strongly associated with the carbanionic centre, its p orbital can even participate in the electronic process allowing processes that would otherwise be symmetry-forbidden.

A plausible structure for **4** may be the α,α-dipotassium derivative of dibenzoheptafulvene (Scheme 3), where a [1,5] hydrogen shift and a [1,2] sigmatropic rearrangement seem to have occurred.

Dibenzopentalene could be another possibility for the final product ([1,2] hydrogen shift; [1,4] sigmatropic rearrangement) but the corresponding radical anion reported in the literature[14] has a very different EPR spectrum. The dibenzopentalene radical trianion has also been considered but not confirmed due to the failure in its direct generation from the parent compound.

In order further to substantiate the proposed structure **4**, the reaction mixture was analysed by HPLC. Two main peaks were observed in the chromatogram. The peak with the larger retention time was assigned to **3** by injection of the reaction mixture with added **3**. The remaining peak could then be assigned to the dibenzoheptafulvene (diprotonated **4**) by an identical method. The much larger retention time of dibenzopentalene, relative to the observed peaks, has ruled out the possibility of this compound as the last species observed.

Once the structure of $\mathbf{4}^{\bullet -}$ is assumed, the question arises as to how the [1,2] migration of a phenyl group can take place when it is known that sigmatropic [1,2] migrations are symmetrically forbidden in neutral molecules and in conjugated anions. A possible answer lies in the intermediacy of **10** which not only defies 'chemical common sense,' but also the Bredt rule. Rather than withhold this, admittedly daring, hypothesis we prefer to submit it for discussion. Further investigation is certainly necessary to substantiate the proposed rationalization of our experimental results.

Experimental

A sample of cycloocta[*b*]naphthalene[15] was provided by Professor H. Günther, University of Siegen. Benzo[*b*]biphenylene,[16] dibenzoheptafulvene[17] and dibenzopentalene[18–20] were synthesized according to standard procedures, and 2-phenylnaphthalene was supplied by Aldrich. MTHF, THF and DME were dried by known techniques.

The reduction of **1–3** with the different alkali metals has been performed by known techniques.

EPR spectra were run on a Bruker ER 200D spectrometer equipped with a variable-temperature unit ER 400 VT. ENDOR and triple-resonance experiments were run on a Bruker EN810 spectrometer. EPR spectra were computer simulated until the match with the experimental spectra could not be further improved.

HPLC product analyses were made with a 10 μm particle size silica gel Lichrosorb Si60 column of 4 mm inner diameter and 250 mm length. Hexane was used as the eluent. A UV detector was tuned to 260 nm.

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