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## Letters

### Clarified charge account

Permit me to all attention to three points in  
Linda Garmon's otherwise excellent article  
("The Disputed Charge Account," SN: 8/13/83, p.  
106).

The classical 2-norbornyl cation does not  
contain the + charge localized at C2. The intro-  
duction of such a charge into the parent hydro-  
carbon, norbornane, will result a) in a flow of  
electrons through the bonds toward the charge  
(inductive effect, C. K. Ingold, 1928), b) an in-  
teraction of the charge through space with sur-  
rounding carbon atoms (field effect, C. K. In-  
gold, 1924), and c) delocalization of charge to  
the adjacent carbon atoms (hyperconjugation,  
R. S. Mulliken, 1939). Consequently, it is uncer-  
tain whether it is the classical or the nonclassi-  
cal structure which delocalizes the + charge  
more effectively from C2 to the rest of the  
molecule.

The essential difference between the  
classical and the nonclassical formulations is  
that of structure. The nonclassical formulation  
possesses a plane of symmetry and elegantly  
accounts for the racemization in the solvolysis

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Cover: Chiclette, the unusually tiny collared lemur born at the Duke Primate Center in April, shares characteristics with human babies whose growth was retarded before birth. She had a relatively large head and little fat, and her mother had previously given birth to small, but not so small, babies. (Photograph: Duke University)
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products from 2-norbornyl derivatives. The classical formulation does not possess a plane of symmetry. It is necessary to postulate a rate of equilibration that is fast compared to the capture of the cation by solvent. Unfortunately, we cannot decide competing scientific explanations on the basis of their relative elegance.

It is an old problem in organic chemistry to decide whether a given system is best described as a rapidly equilibrating pair (double minima) or a single intermediate structure (single minimum). Fifty years ago I was taught that benzene was a double minima system, whereas the hydrogen bond and cyclohexane were single minimum systems. Now our position in these three cases is the exact opposite.

The nonclassical formulation was also used to explain the high exo/endo rate ratios in the solvolysis of 2-norbornyl derivatives. This has a precise quantitative requirement. The transition state for secondary exo/norbornyl derivatives must be stabilized by six kcal of energy and a somewhat greater quantity in the free ion, a stabilization that is absent in tertiary 2-norbornyl. But all of the data show the absence of this nonclassical stabilization energy. The differences in energy between secondary

and tertiary cations are the same for representative aliphatic cations, monocyclic cations and 2-norbornyl cations. None of the supporters of the nonclassical formulation has yet addressed this finding.

The precise nature of the 2-norbornyl cation in the gas phase or in the solid state at very low temperatures is an interesting question, but does not provide a quantitative basis for the high exo/endo rate ratio under solvolytic conditions, responsible for the original nonclassical proposal.

Finally, I believe the article does not do justice to the effectiveness of my reexamination of the nonclassical question. In their heyday, nonclassical structures were considered for essentially all carbonium ions known to man other than methyl,  $\text{CH}_3^+$ . Now essentially all are assigned classical structures. We are presently discussing 2-norbornyl as a residual case to be decided. The fact that my campaign has brought chemists 99% of the way toward the classical formulation for solvated carbocations does not come through your story.

Herbert C. Brown  
Dept. of Chemistry, Purdue Univ.  
W. Lafayette, Ind.

