Table II. Calculated Absolute NMR Shieldings[a]

	7-oxa[2.2.1]hericene (**2**)			tetrakis(methylene)bicyclo[2.2.0]hexane (**3**)			*o*-xylylene (**14**)		
basis	energy[b]	X	Y	energy	X	Y	energy	X	Y
6-31G**	−479.153 92	64.04	98.26	−384.371 44	56.77	98.03	−307.553 27	66.02	91.43
6-31+G**	−479.168 94	60.83	100.21	−384.382 94	53.67	100.37	−307.563 57	63.65	93.74
6-311G**	−479.250 62	47.19	86.12	−384.439 60	40.94	86.08	−307.608 44	51.35	79.57

[a] Calculated with the GIAO method at the SCF/6-31G** optimized geometries; unit is ppm. [b] Total SCF energy; unit is hartree.

both the NMR and UV–vis signals. However, the possibility that the new NMR signal originates from 2,3,5,6-tetrakis(methylene)bicyclo[2.2.0]hexane (**3**) was not excluded completely, although the explanation of the UV–vis signal seems less satisfactory in the latter case.[5]

To help clarify the identity of the photoproduct, we carried out theoretical calculations on the structures and magnetic shielding tensors for the vinyl carbons X and Y of **2**, **3**, and *o*-xylylene (**4**). It would be more conclusive to calculate the NMR shieldings of TMB, too; however, the program for open-shell molecules is not available at present. Nevertheless, on the basis of the structural and electromagnetic environmental similarities, it is reasonable to expect that the chemical shift of the methylene carbon of **4** is close to that of the singlet TMB.

All the structures were fully optimized at the SCF level using the 4-21G[15] and 6-31G**[16a] basis sets. The results for **2** and **3** are given in Table I. Note that the $C_2C_1C_6$ bending and $C_2C_1C_4C_5$ dihedral angles of **3** vary significantly (≈3°) with basis sets. This indicates that the molecule is highly strained. Since higher angular momentum polarization functions give better descriptions for strained molecules, the results obtained with the 6-31G** basis are expected to be more reliable. Consequently, the NMR shielding calculations were carried out at the 6-31G** geometries by the gauge-independent atomic orbital (GIAO) method[17] at the SCF level using the 6-31G**, 6-31+G**, and 6-311G** basis sets.[16] The program TX90[18] was used for all the calculations.

Table II contains the calculated absolute NMR shieldings and SCF energies. With all three basis sets, the methylene carbon shieldings of **2** and **3** differ by less than 1 ppm. This indicates that if **3** were produced in the photolysis of **2**, its methylene ^{13}C signal would be indistinguishable from the precursor signal. On the other hand, the NMR shielding of the methylene carbon of **4** is calculated to be lower than that of **2** by 6.83 (6-31G**), 6.47 (6-31+G**), and 6.55 (6-311G**) ppm. This difference corresponds well with the observed signal of the photoproduct of Berson et al. Therefore, it is reasonable to believe that the photoproduct is in a singlet state and has a structural unit similar to **4**.

Presumably due to the high reactivity, there have been no reports of ^{13}C NMR measurements for **3** and **4**. For **2**, the ^{13}C NMR shifts of atoms X and Y are reported[19] to be 142.5 and 103.8 ppm, respectively. The difference, 38.7 ppm, agrees very well with our calculated results, 34.22 (6-31G**), 39.38 (6-31+G**), and 38.93 (6-311G**) ppm. In fact, in previous studies[17b] in this laboratory, a chemical shift difference as small as 0.7 ppm between the para and meta carbons in styrene has been correctly reproduced by using the 6-31G(d) basis set.

In summary, this theoretical study provides convincing evidence supporting the conclusion of Berson et al. that the new ^{13}C NMR signal observed at 113 ppm is not that of **3**. The fact that 2,3,5,6-tetrakis(methylene)bicyclo[2.2.0]hexane was not produced in the photolysis of 7-oxa[2.2.1]hericene may be explained by the calculated molecular structures. To produce **3**, the distance between C_1 and C_4 has to decrease from 2.30 Å to 1.57 Å, which invokes a strong strain effect, and therefore, this mechanism is unfavorable kinetically.

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Intracluster Ion–Molecule Reactions within Ethylene–Methanol Heteroclusters

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Recent studies on the properties, structure, and reactivities of molecular clusters contribute to an understanding of the reaction dynamics lying between the gaseous and condensed phases.[1-3] Apart from the numerous investigations of the structure of the ions formed in gas-phase ion–molecule association reactions, there are few reported cases of chemical reactions taking place within the heterocluster ion itself.[4-6]

In this study we have investigated the $(C_2H_4)_n(CH_3OH)_m$ heterocluster systems to look for new intracluster ion–molecule reactions occurring within the ionized clusters and to elucidate reactive pathways not ordinarily found in bimolecular ion–molecule collisions. The molecular beam apparatus used in this work has been described previously.[7] Neutral heteroclusters are generated by expanding a gas mixture (6% C_2H_4; 0.03–0.6% CH_3OH) seeded in 4.5 atm of He through an 800-μm pulsed nozzle. Ions formed by the electron impact are accelerated in a double electrostatic field to 1.1 keV and directed through a 1-m-long flight tube. The mass spectrum is recorded by a transient digitizer coupled with a microcomputer.

Figure 1 displays the mass spectra taken at different mixing ratios of ethylene/methanol (E/M) of 10/1 and 100/1. Though $E_nM_mH^+$ ions show only small contributions compared to the corresponding unprotonated species in the 10/1 ratio, the same ions are now substantially increased in intensity when they are formed at the 100/1 ratio. This observation is easily explained assuming that the proton is bound to a methanol molecule in $E_nM_mH^+$ ions due to the large difference between the proton affinities[8] of CH_3OH (8.03 eV) and C_2H_4 (7.10 eV). A similar result has also been found in a study of the dissociation of mixed ammonia–acetone cluster ions.[9,10] In the 10/1 ratio, the pro-

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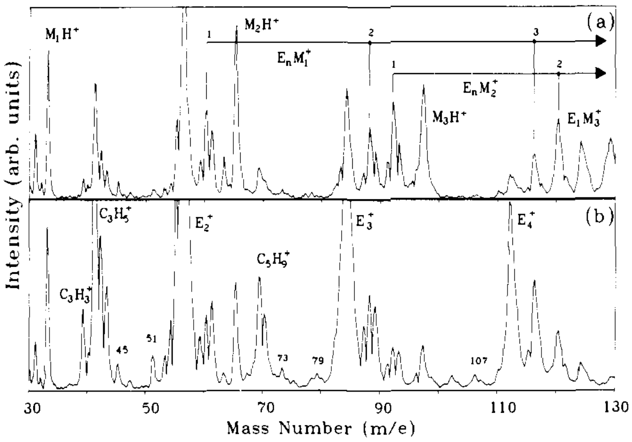


Figure 1. Comparison of electron-impact (50 eV) TOF mass spectra of mixed ethylene–methanol clusters formed at the different mixing ratios: (a) C_2H_4 (6%) + CH_3OH (0.6%); (b) C_2H_4 (6%) + CH_3OH (0.06%). Integers above the peaks denote *n* and *m*, the numbers of C_2H_4 and CH_3OH molecules in an individual cluster ion, respectively. E and M designate C_2H_4 and CH_3OH molecules.

tonated methanol ion in the heterocluster will be preferentially solvated by the neutral methanol molecules, since hydrogen bonding is not available to ethylene molecules. This situation is similar to that of the ion–molecule reactions occurring in the methanol homoclusters, resulting in only a minor contribution from protonated heterocluster ions. When the mixing ratio is increased to 100/1, however, the preferential solvation of protonated methanol ion by the neutral methanol molecules is now unfavorable because of the extremely low methanol concentration. For this reason, protonated methanol ion would presumably have a greater chance to react with the ethylene molecules, thereby showing the increased prominence of $E_nM_mH^+$ ions.

Another distinct feature is that $E_nM_mH^+$ ions make a substantial contribution only when $n + m \leq 3$. This finding is predominant over a wide range of vapor composition, indicating that $E_nM_mH^+$ ions with $n + m \leq 3$ have particularly stable structures. Hiraoka and Kebarle[11] have reported that the reaction between CH_3OH and alkyl carbocation in solution leads to the protonated methyl alkyl ether. Formation of $[(C_2H_4)(CH_3OH)]H^+$ ion can be interpreted as a result of an intracluster ion–molecule condensation reaction, which produces a protonated methyl ethyl ether.[12] In view of the preceding results of the intensity dependence of protonated heteroclusters on the mixing ratios, one may conclude that reaction 1 prevails against 2.

$$[CH_3OH_2^+ + C_2H_4]E_nM_m \rightarrow CH_3O^+(H)C_2H_5 + nE + mM \tag{1}$$

$$[CH_3OH + C_2H_5^+]E_nM_m \rightarrow CH_3O^+(H)C_2H_5 + nE + mM \tag{2}$$

Around a central $CH_3O^+(H)C_2H_5$ ion there is room for one hydrogen-bonded methanol molecule to form a $[(C_2H_4)(CH_3OH)_2]H^+$ ion. Higher cluster ions, $[(C_2H_4)(CH_3OH)_m]H^+$ ($m \geq 3$), however, reflect the incorporation of the hydrogen-bonded methanol chains to the protic sites on the core ion. This is primarily attributed to the large binding energy change that can be caused when the first solvent shell is filled by ligands with an alkyl blocking group.[13]

In order to explain the observed $[(C_2H_4)_2(CH_3OH)]H^+$ ion, the following reaction is proposed:

$$CH_3O^+(H)C_2H_5E_nM_m \rightarrow [CH_3O^+(H)C_2H_5 + C_2H_4]E_{n-1}M_m \rightarrow CH_3O^+(C_2H_5)_2 + (n-1)E + mM \tag{3}$$

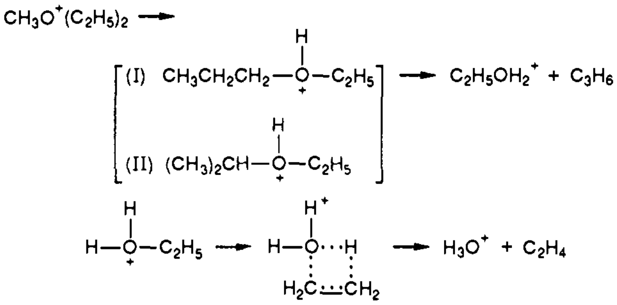
Table I. Observed Intensity Ratios at Different Mixing Conditions

$[C_2H_4]/[CH_3OH]$ ratio	$E_1M_1H^+/C_2H_5O^+$	$E_1M_1H^+/E_1M_1^+$	$E_2M_1H^+/E_2M_1^+$	$E_1M_2H^+/E_1M_2^+$
200/1	3.60	1.36	0.82	1.14
100/1	3.07	1.16	0.90	1.00
50/1	3.04	0.86	0.72	0.68
30/1	3.00	0.67	0.63	0.61
10/1	3.11	0.47	0.61	0.59

The ion–molecule reaction of two ethylene molecules with a protonated methanol, within the bulk cluster, forms a tertiary oxonium intermediate via $CH_3O^+(H)C_2H_5$. Minor contributions from $E_nM_mH^+$ ($n \geq 3$ and $m \geq 2$) ions indicate that either hydrogen bonding by a CH_3OH molecule or the association reaction of C_2H_4 to oxonium ion is not available.

It is worthy of note that $C_2H_5O^+$ ion (mass 45) is only salient as the intensities of the protonated heterocluster ions are increased. The observation of $C_2H_5O^+$ intensity representing the same propensity (see Table I) with $E_1M_1H^+$ [$\equiv CH_3O^+(H)C_2H_5$] implies the formation of $CH_2{=}O^+CH_3$ ion. Harrison et al.[14] reported that the ion formed from methyl ethyl ether should have the structure $CH_2{=}O^+CH_3$. Now one might expect that the protonated dimethyl ether ion, produced via a dehydration reaction in the protonated methanol dimer ion, could undergo H_2 elimination to yield $CH_2{=}O^+CH_3$. However, the total absence of this peak in the mass spectrum of pure methanol excludes this process.

Another finding of particular importance is the observation of a peak corresponding to $(CH_3OH)H_3O^+$ (mass 51) ion.[15] It is interesting to note that while $(C_2H_4)(CH_3OH)H^+$ and $(C_2H_4)(CH_3OH)_2H^+$ ion intensities relative to their corresponding unprotonated species increase with the C_2H_4/CH_3OH ratio, the increasing tendency of $(C_2H_4)_2(CH_3OH)H^+$ is substantially reduced at a 200/1 ratio (see Table I). We speculate that the formation of H_3O^+ can be attributed to $(C_2H_4)_2(CH_3OH)H^+$ ion being consumed by an intracluster ion–molecule rearrangement following fragmentation:



$$CH_3O^+(C_2H_5)_2 \rightarrow \left[\begin{array}{l} \text{(I)}\ CH_3CH_2CH_2{-}\overset{H}{\underset{+}{O}}{-}C_2H_5 \\ \text{(II)}\ (CH_3)_2CH{-}\overset{H}{\underset{+}{O}}{-}C_2H_5 \end{array}\right] \rightarrow C_2H_5OH_2^+ + C_3H_6 \tag{4}$$

$$H{-}\overset{H}{\underset{+}{O}}{-}C_2H_5 \rightarrow H{-}\overset{H^+}{O}\cdots H \ (H_2C{\cdots}CH_2) \rightarrow H_3O^+ + C_2H_4 \tag{5}$$

Recently, this type of ion–molecule rearrangement reaction has been studied by Coolbaugh et al.[16] upon the electron impact of the neutral dimethyl ether clusters. Within stabilizing environs of a cluster, the intermediate may now dissociate internally, where it then forms protonated ethanol via elimination of propylene. Direct evidence for the formation of ion II comes from the observation of a mass 73 peak in Figure 1b. Bowen and Maccoll[17] have observed the α-cleavage fragment $CH_3C^+HOC_2H_5$ ion (mass 73) from isopropyl ethyl ether using low-energy and low-temperature mass spectrometry.

The formation of H_3O^+ by a β-hydrogen-transfer process[18,19] has a lower reaction enthalpy and begins to dominate over the formation of $C_2H_5^+$ at low collision energies. The increased

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intensity of mass 51 ion at a high C_2H_4/CH_3OH ratio suggests that ethylene molecules have the greater opportunity to react with the protonated methanol ion in the presence of trace methanol. Thereafter the solvating ethylene molecules form the observed $(C_2H_4)_n(CH_3OH)H_3O^+$ ions. This sequence of peaks is observed throughout the entire cluster mass spectrum to the limit of our sensitivity.

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Registry No. Ethylene, 74-85-1; methanol, 67-56-1.

Push-Pull Porphyrins as Nonlinear Optical Materials

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Due to their potential applications for optical communications, data storage, and electrooptical signal processing, molecules with nonlinear optical (NLO) properties have been extensively investigated.[1] As part of our interest in porphyrins and metalloporphyrins as photoresponsive[2] and field-responsive materials,[3] we have synthesized a series of "push-pull" porphyrins containing both donor and acceptor substituents and have examined their NLO properties. The enhancements in molecular hyperpolarizabilities in previous organic systems have relied on donor–acceptor interactions along one-dimensional pathways; our porphyrinic compounds are prototypes of donor–acceptor interactions in two dimensions.

Porphyrins with their large π-conjugated system provide a versatile synthetic base to create effective intramolecular charge transfer (CT). As shown in Figure 1, the difunctionalized tetraarylporphyrins with nitro groups (n) as electron acceptors and amino groups (a) as electron donors have been prepared. The synthesis of H_2(an$_3$Por), H_2(*cis*-a$_2$n$_2$Por), and H_2(a$_3$nPor) does not involve the traditional crossed-condensation for multisubstituted porphyrins,[4] which are usually isolated only in low yields (<5%). Instead, H_2(n$_4$Po4), H_2(an$_3$Por), H_2(*trans*-a$_2$n$_2$Por), H_2(*cis*-a$_2$n$_2$Por), H_2(a$_3$nPor), and H_2(a$_4$Por) were prepared by the partial reduction of the nitro groups of 5,10,15,20-tetrakis(*p*-nitrophenyl)porphyrin[5] with 2.5 times the stoichiometric amount of $SnCl_2$. The reaction was performed under nitrogen in concentrated hydrochloric acid at 75 °C for 24 h. After the reaction was neutralized with concentrated ammonium hydroxide, the isolated solid mixture was placed in a Soxhlet apparatus and extracted with chloroform for 7 days. H_2(an$_3$Por), H_2(*cis*-a$_2$n$_2$Por), and H_2(a$_3$nPor) were separated from each other by column chromatography in ~30% isolated yields each. They have been fully characterized by elemental analysis, TLC, and FAB-MS, UV–visible, FTIR, and ^{1}H NMR spectroscopies, as provided in the supplementary material. The optical spectra of these porphyrins are included in the supplementary material and show no absorbances at wavelengths greater than 680 nm.

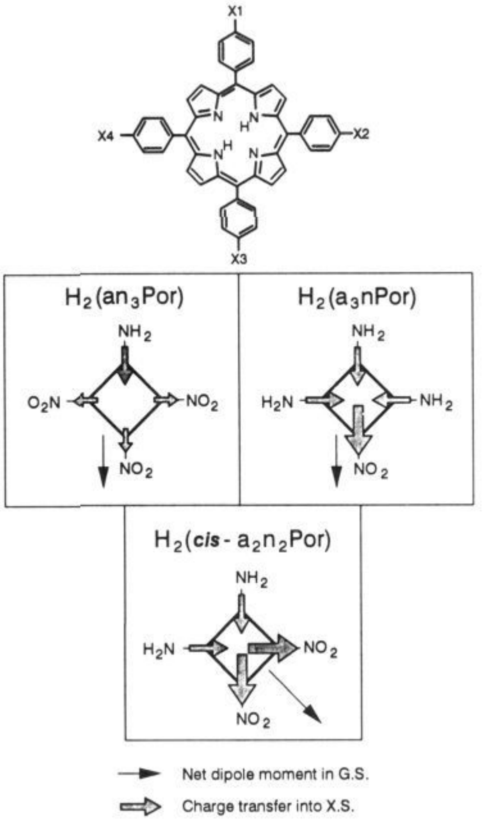

Figure 1. Structure, net dipole moment vectors, and charge-transfer transition vectors of push-pull porphyrins: H_2(n$_4$Por), X1 = X2 = X3 = X4 = NO_2; H_2(an$_3$Por), X1 = X2 = X3 = NO_2, X4 = NH_2; H_2(*trans*-a$_2$n$_2$Por), X1 = X3 = NO_2, X2 = X4 = NH_2; H_2(*cis*-a$_2$n$_2$Por), X1 = X2 = NO_2, X3 = X4 = NH_2; H_2(a$_3$nPor), X1 = NO_2, X2 = X3 = X4 = NH_2; H_2(a$_4$Por), X1 = X2 = X3 = X4 = NH_2; H_2TPP, X1 = X2 = X3 = X4 = H.

The structural control of β, the second-order molecular hyperpolarizability, is well understood from both theoretical and experimental perspectives.[6] Organic molecules with strong electron donor and electron acceptor groups that are connected by a large conjugated π-electron system usually show high β values. Following this general rule, a variety of stilbenes, polyenes, and diazo molecules have been designed that have β values up

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