Internet Electronic Journal of Molecular Design **2002**, *1*, 52–58 ISSN 1538–6414
BioChem Press http://www.biochempress.com

Internet *net* Electronic Journal of
Molecular Design

January 2002, Volume 1, Number 1, Pages 52–58

Editor: Ovidiu Ivanciuc

Special issue dedicated to Professor Alexandru T. Balaban on the occasion of the 70th birthday
Part 1

Guest Editor: Mircea V. Diudea

There Are No Antiaromatic Molecules in Interstellar Space

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Received: November 13, 2001; Accepted: December 15, 2001; Published: January 31, 2002

Citation of the article:
J. Aihara and M. Hirama, There Are No Antiaromatic Molecules in Interstellar Space, *Internet Electron. J. Mol. Des.* **2002**, *1*, 52–58, http://www.biochempress.com.

Inter*net* Electronic Journal of Molecular Design

BioChem Press
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There Are No Antiaromatic Molecules in Interstellar Space[#]

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Internet Electron. J. Mol. Des. **2002**, *1* (1), 52–58

Abstract

Motivation. Some unsaturated interstellar molecules are cyclic in geometry. We aimed at estimating the degree of aromaticity in these molecules.

Method. π–Electrons in 14 ring molecules were enumerated using the B3LYP/6–31G** level of theory. TREs were then calculated for them.

Results. All interstellar ring molecules with π–electrons and their derivatives were found to be highly aromatic with large positive TREs.

Conclusions. It seems quite likely that antiaromatic species are scarcely formed in interstellar space. This aspect of interstellar chemistry will be useful for designing or predicting unknown interstellar molecules.

Keywords. Interstellar molecule; topological resonance energy; aromaticity; antiaromaticity.

Abbreviations and notations

HOMO, highest occupied molecular orbital	TCS, topological charge stabilization
MO, molecular orbital	TRE, topological resonance energy
PAH, polycyclic aromatic hydrocarbon	

1 INTRODUCTION

More than 120 molecules have now been identified in interstellar space and/or circumstellar shells [1,2]. Most of the interstellar molecules with more than three atoms are organic in nature. It is noteworthy that highly unsaturated carbon compounds are so conspicuous in space where hydrogen, by a very large factor, is the most abundant chemically active element [2]. Gas–phase ion–molecule reactions are believed to be primarily responsible for the formation of such interstellar molecules [1] although much more needs to be learned about their mechanisms.

We have been interested in structural aspects of interstellar molecules and found that they in principle abide by the TCS rule [3]. This rule states that nature prefers to locate electronegative heteroatoms at sites that conform to the pattern of relative electron densities determined by

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[#] Dedicated on the occasion of the 70th birthday to Professor Alexandru T. Balaban.

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connectivity or topology in an isoelectronic, isostructural, homoatomic system [4]. Chemically stable interstellar molecules, such as CO_2, OCS, HNCO, NH_2CHO, HCOOH, and $HCOOCH_3$, are fully consistent with the TCS rule [3].

Some unsaturated interstellar molecules are cyclic in geometry [2]. Therefore, there is a possibility that aromaticity [5] might also govern the formation of interstellar ring molecules. The TRE has been used as a simple but practical index for determining the degree of aromaticity [6,7]. However, it is not easy to calculate TREs for interstellar molecules because one cannot readily enumerate π–electrons they have. All known interstellar ring molecules are rather exotic free radicals or carbenes. We carried out density–functional calculations to determine the number of π–electrons in interstellar ring molecules and related species, then calculated the TREs, and examined the role of aromaticity in interstellar chemistry.

2 METHODS

Ab initio or density–functional calculations are necessary to investigate electronic structure of free radicals and carbenes. In this study, the number of π–electrons and the MO energies for ring molecules **1**–**14** in Figure 1 were determined at the B3LYP/6–31G** level of theory using the Gaussian 98 program package [8]. Fortunately, it has been established that **2**–**4**, **6**–**8**, and **10**–**13** have a closed–shell singlet ground state [9–19], whereas **1**, **5**, and **9** are in the doublet electronic state [20–23]. Pentagonal carbene **14** alone has a triplet ground state [24,25]. We assumed such spin states in MO calculations. Spin contamination does not appear to be a problem at the present level of theory. The expectation values of S^2 were 0.754 for **1**, 0.759 for **5**, 0.767 for **9**, and 2.023 for **14**, thus being very close to nominally exact values. For open–shell species, the energy of every π–MO was obtained by averaging it over the two different spin states. TREs were calculated using Van–Catledge's Hückel parameters for heteroatoms [26].

3 RESULTS AND DISCUSSION

The number of π–electrons, energies of π–MOs, and TREs calculated for **1**–**14** are listed in Table 1, together with the energies of HOMOs. Of these ring molecules, **1**–**3** and **13** have been detected spectroscopically in astronomical sources [20,27–29]. The other molecules have been proposed as candidates for astronomical detection [2,30,31]. Most of them must be very reactive at terrestrial densities because one or more constituent atoms do not satisfy the octet rule. In line with this, the HOMOs in **1**–**13** are not π–MOs.

Interstellar molecules C_3H (**1**), C_3H_2 (**2**), and SiC_2 (**3**) have two π–electrons in their three–membered rings [9,10,21]. These molecules are highly aromatic even if they are very reactive in the laboratory. Their TREs are positive in sign and much larger than those for benzene (0.273 |β|) and for naphthalene (0.389 |β|) [6,7]. Here, β is the standard resonance integral for carbon–carbon π bonds. The TRE of 0.273 |β| for benzene corresponds to the Dewar resonance energy of ca. 20 kcal/mol [32]. The π–MOs in **1**–**3** lie as low in energy as those in typical benzenoid hydrocarbons. Note that the HOMOs in benzene and naphthalene are located at –9.24 and –8.12 eV, respectively [33]. Thus, it is reasonable to presume that two π–electrons in **1**–**3** contribute much to the stabilization of the molecule. Silicon compound **3** has one more π–like MO with energy –101.01 eV. This MO must contribute little to aromaticity because it is localized at the silicon $2p_z$ atomic orbital.

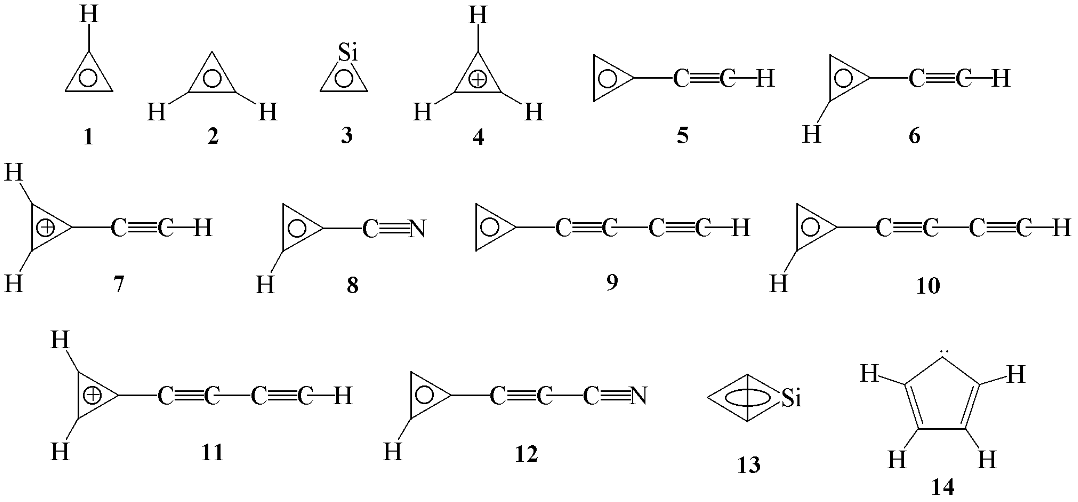

Figure 1. Interstellar ring molecules and related species studied.

Table 1. Numbers of π–electrons, Energies of the HOMO and π–MOs, and TREs for Compounds **1**–**14**

Specie	Number of π–electrons	Energy of the HOMO / eV	Energies of π–MOs / eV	TRE / |β|
C_3H (**1**)*	2	–6.87	–10.66	0.536
C_3H_2 (**2**)*	2	–6.09	–10.22	0.536
SiC_2 (**3**)*	2	–7.06	–9.04	0.430
$C_3H_3^+$ (**4**)	2	–17.39	–18.04	0.536
C_5H (**5**)	4	–6.85	–11.28, –8.70	0.429
C_5H_2 (**6**)	4	–6.13	–10.92, –8.28	0.429
$C_5H_3^+$ (**7**)	4	–13.35	–17.62, –13.88	0.429
C_4NH (**8**)	4	–7.05	–11.95, –9.83	0.471
C_7H (**9**)	6	–6.51	–11.42, –10.15, –7.90	0.376
C_7H_2 (**10**)	6	–6.11	–11.11, –9.79, –7.53	0.376
$C_7H_3^+$ (**11**)	6	–11.74	–17.05, –14.54, –12.19	0.376
C_6NH (**12**)	6	–6.82	–11.87, –10.90, –8.69	0.420
SiC_3 (**13**)*	2	–6.34	–10.65	0.759
C_5H_4 (**14**)	5	–6.12	–9.91, –6.12, –6.12	–0.301

* Interstellar molecules.

$C_3H_3^+$ (**4**) is the simplest aromatic annulene ion and is familiar as the mass 39 peak in mass spectra [31]. This classical carbocation likewise has two π–electrons [12]. It follows that three–membered rings **1**–**4** are iso–π–electronic with each other. If they had one more π–electron, three in all, they would be highly antiaromatic with a large negative TRE of –0.464 |β|. Cation **4** is non–polar by symmetry and possesses no microwave spectrum. However, this ion is almost certainly present in interstellar space, where it is thought to be the precursor of **2** [31]. Since **4** is positively charged, even the HOMO is much lower in energy than those in neutral species.

So far hypothetical interstellar molecules have been proposed in relation to **1**–**4**. In particular, ring–chain molecules such as **5**–**12** are noteworthy [2,31] because many highly unsaturated carbon chains are detectable in space. It has then been argued that these ring–chain molecules are stabilized owing to the presence of 4n+2 (n=0) π–electrons in the ring [15–17,34]. All of them in fact are highly aromatic with large positive TREs, having enough π–electrons to maximize the TRE. Ring–chain cations **7** and **11** are supposed to be the precursors of **6** and **10**, respectively, in space [31]. Since they are positively charged, even the HOMOs lie very low in energy. Thaddeus et al. detected **6**, **8**, **10**, and **12** by laboratory experiments [34–36]. No one has ever observed **5**, **7**, **9**, and **11** in space or in the laboratory [31].

Rhomboidal SiC_3 (**13**) is not only the largest ring molecule so far identified in space but also the only bicyclic interstellar molecule [29]. This molecule has two π–electrons in a low–lying MO [18], which suffice for attaining the maximum degree of aromaticity. Such high aromaticity is presumably responsible for its planar geometry and the singlet electronic state. As in the case of **3**, there is one more π–like orbital in **13**. It, however, contributes little to aromaticity since it is localized at the silicon $2p_z$ atomic orbital.

Oka et al. detected the H_3^+ ion in many celestial objects [37]. This triatomic ion is the precursor for much of the gas–phase chemistry that occurs in dense interstellar clouds [1]. This ion has no π–electrons. However, it can be considered as an aromatic species since two electrons are delocalized in the three–membered ring. It is necessarily isoelectronic with the π–electronic systems of **1**–**4**. If this ion were antiaromatic, it would not form in space. Thus, we might safely say that the aromaticity of H_3^+ is the origin of the entire interstellar chemistry.

In addition to fairly simple molecules, much larger aromatic molecules are known to exist abundantly in space. In 1984 Léger and Puget noticed that there are a mixture of large PAH molecules and their molecular ions in many celestial objects, such as planetary nebulae, reflection nebulae, H II regions, and other galaxies [38,39]. Furthermore, the hypothesis that most of the intense diffuse interstellar bands are due to a mixture of ionized PAHs has gained some plausibility although convincing laboratory evidence is not yet available [40]. PAHs are very stable aromatics with large positive TREs [41].

Here, we should note that not only aromatic molecules but also high–energy isomers of some stable molecules are among interstellar molecules [2,31]. For example, vinyl alcohol $CH_2{=}CHOH$ is ca. 15 kcal/mol less stable than the structural isomer acetaldehyde CH_3CHO, both being interstellar molecules [42]. Therefore, it is very strange that antiaromatic molecules, a kind of high–energy species, have not been detected in space. Why not? Unfortunately, we don't have a definite answer to this question. It may be simply related to the fact that many interstellar ring molecules have not been detected yet. Formation of strained three–membered rings, such as **1**–**3** and **13**, might be the result of effort with which the constituent atoms try to satisfy the octet rule. However, it is certainly true that they are greatly stabilized by a large aromatic stabilization energy.

Cyclopentadienylidene C_5H_4 (**14**) is a carbocyclic carbene homologous to cyclopropenylidene (**2**). In marked contrast to interstellar ring molecules so far detected, this ring molecule has a triplet ground state with five π–electrons and proved to be antiaromatic with a large negative TRE. Considering that all known interstellar molecules are never antiaromatic, it does not seem likely that **14** is detectable in space. In general, a monocyclic system with an odd number of π–electrons is antiaromatic [6,7]. The microwave detection of **14**, if any, might be complicated somewhat by its rich spectrum and rather small dipole moment [30]. The HOMO in **14** is a π–MO, which lies high at –6.12 eV.

4 CONCLUSIONS

Interstellar ring molecules so far detected were found to be aromatic with large positive TREs. This tempts us to suggest that interstellar chemistry tends to generate non–antiaromatic molecules for some kinetic reasons. Therefore, when one designs novel interstellar molecules, it is very desirable that they have no antiaromatic character. In fact, there is one more interstellar ring molecule, oxirane C_2H_4O [43]. It is a fully saturated stable molecule and therefore was not considered in this paper.

We previously pointed out that biomolecules obey the TCS rule [44] and that there are no antiaromatic biomolecules [45,46]. Although some organisms living on Earth can produce highly strained molecules with many cyclopropyl rings, they wouldn’t produce antiaromatic molecules [47]. Nature prefers strained molecules to antiaromatic ones. Thus, it is interesting to see that similar situations arise in the interstellar medium.

Acknowledgment

This work was supported by a Grant–in–Aid for Scientific Research (No. 11640502) from the Ministry of Education, Science, Sports, and Culture, Japan. We are grateful to Prof. Shigeru Ikuta (Tokyo Metropolitan University) and Prof. Ken Sakata (Hoshi College of Pharmacy) for helpful and instructive suggestions. Computation was performed at the Information Processing Center, Shizuoka University, and the Computer Center, Institute for Molecular Science.

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