# Aromaticity, Antiaromaticity, Homoaromaticity and the Hückel (4n + 2) Rule

Present	entation · December 2018			
DOI: 10.131	.13140/RG.2.2.34131.63528			
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## **\*** Aromaticity

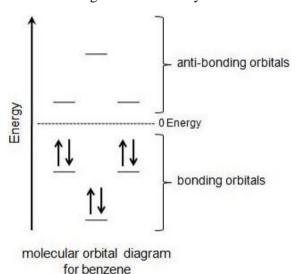
- In 1931, German chemist and physicist *Sir Erich Hückel* proposed a theory to help determine if a planar ring molecule would have aromatic properties. His rule states that if a cyclic, planar molecule has  $4n+2\pi$  electrons, it is considered aromatic. This rule would come to be known as *Hückel's Rule*.

### • Criteria for Aromaticity

- 1) The molecule is cyclic (a ring of atoms)
- 2) The molecule is planar (all atoms in the molecule lie in the same plane)
- 3) The molecule is fully conjugated (**p orbitals** at every atom in the ring)
- 4) The molecule has  $4n+2\pi$  electrons (n=0 or any positive integer)

### • Why $4n+2 \pi$ Electrons?

- According to Hückel's Molecular Orbital Theory, a compound is particularly stable if all of its bonding molecular orbitals are filled with paired electrons.
- This is true of aromatic compounds, meaning they are quite stable.
- With aromatic compounds, **2 electrons** fill the lowest energy molecular orbital, and **4 electrons** fill each subsequent energy level (the number of subsequent energy levels is denoted by n), leaving all bonding orbitals filled and no anti-bonding orbitals occupied. This gives a total of **4n+2** $\pi$  electrons.
- As for example: Benzene has  $6\pi$  electrons. Its first  $2\pi$  electrons fill the lowest energy orbital, and it has  $4\pi$  electrons remaining. These 4 fill in the orbitals of the succeeding energy level. Notice how all of its bonding orbitals are filled, but none of the anti-bonding orbitals have any electrons.



- To apply the 4n+2 rule, first count the number of  $\pi$  electrons in the molecule. Then, set this number equal to 4n+2 and solve for n. If is 0 or any positive integer (1, 2, 3,...), the rule has been met.



$$4n + 2 = \pi$$

$$4n + 2 = 6$$

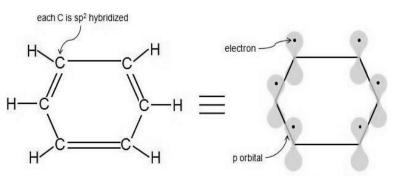
$$4n + 2 = 6$$

$$4n = 6 - 2$$

$$4n = 4$$

$$n = 4/4$$

$$n = 1$$



benzene has 6  $\pi$  electrons

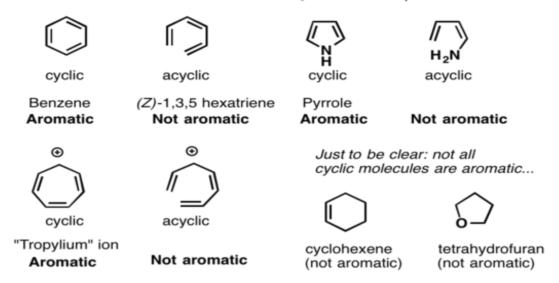
For benzene, we find that n = 1, which is a positive integer, so the rule is met.

Molecules that have the 3 characteristics listed above (*cyclic*, *conjugated*, *flat*) and have this number of  $\pi$  electrons [4n +2] will be aromatic. The letter " $\mathbf{n}$ " is not a characteristic of the molecule because " $\mathbf{n}$ " comes from algebra, NOT from chemistry.

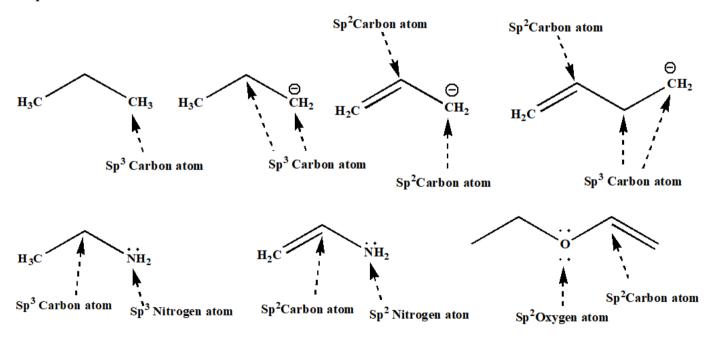
### (i) Condition - 1: The Molecule Must Be Cyclic

- Determining if a molecule is cyclic is pretty straightforward. Then, move to condition -2. If there's no ring, forget it.
- Because as for example: (Z)-1,3,5 hexatriene has the same number of pi bonds (and pi electrons) as benzene, but isn't aromatic. No ring, no aromaticity.

Condition #1: The molecule must be cyclic No exceptions!



- **Note By:** If an Atom has 1 or **more lone pair electron** and is attached to an  $sp^2$  atom then this **atom** is also  $sp^2$  atom.



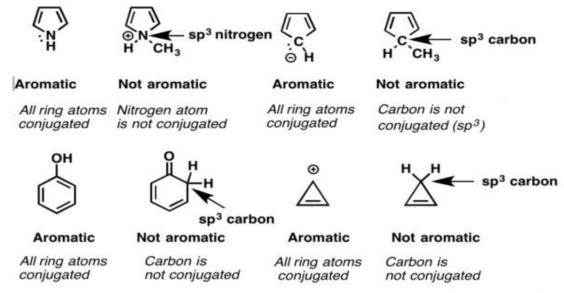
#### (ii) Condition - 2: Every atom in the ring must be conjugated

- Obviously, being cyclic isn't a sufficient condition for aromaticity.
  - "Every atom in the ring must have an available p orbital", or
  - o "Every atom in the ring must be able to participate in resonance".
- In order for aromaticty to exist, there must also be a continuous ring of p-orbitals around the ring that build up into a larger cyclic "pi  $(\pi)$  system".
- Remember that the "available p orbital" condition applies not just to atoms that are part of a pi  $(\pi)$  bond, but also atoms bearing a lone pair, a radical, or an empty p orbital (e.g. carbocations).
- **NOTE BY:** The key thing that "**kills**" conjugation is a  $sp^3$  **hybridized** atom with four bonds to atoms. Such an atom cannot participate in resonance.

#### Condition #2: Every Atom In The Ring Must Be Conjugated

(alternative): every atom in the ring must have an available p orbital

(alternative): every atom in the ring must be able to participate in resonance



an atom attached to four bonds e.g. sp3 carbon, or nitrogen (Not aromatic)

#### (iii) Condition - 3: The Molecule Must Have [4n+2] Pi $(\pi)$ Electrons

- The third condition is that the cyclic, conjugated molecule must have the correct number of **pi**  $(\pi)$  electrons.
- Benzene and Cyclooctatetraene are both cyclic and conjugated, but <u>benzene</u> is aromatic and Cyclooctatetraene is not. The difference is that **benzene** has 6 pi  $(\pi)$  electrons, and Cyclooctatetraene has 8 pi  $(\pi)$  electrons.
- 4n+2 are not a formula which applies to see the molecule is aromatic. It is a formula that tells what numbers are in the magic series. If **pi**  $(\pi)$  **electron** value matches any number in this series then they have the capacity for **aromaticity**."
- The "magic series" is: 2, 6, 10, 14, 18, 22.....

So for n = 0, we have [4(0) + 2] = 2

for n = 1, we have [4(1) + 2] = 6

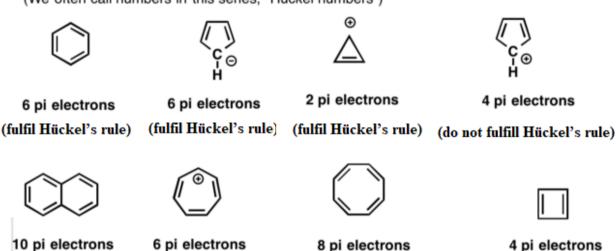
for n = 2, we have [4(2) + 2] = 10

for n = 3, we have [4(3) + 2] = 14

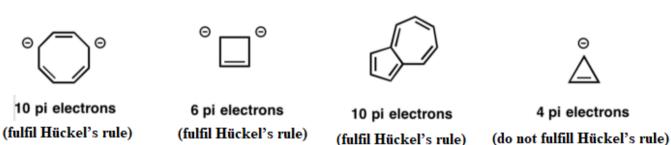
- The condition that aromatic molecules must have [4n+2] pi  $(\pi)$  electrons is sometimes called "Huckel's rule".

#### Condition #3: The Molecule Must Contain [4n + 2] Pi electrons \*

- where [4n+2] is a formula describing the number series: 2, 6, 10, 14, 18, 22, 26....
- These are "magic numbers" for aromaticity: the number of pi electrons in the molecule **must be in this series** in order for the molecule to be aromatic (We often call numbers in this series, "Huckel numbers")



(fulfil Hückel's rule) (fulfil Hückel's rule) (do not fulfill Hückel's rule) (do not fulfill Hückel's rule)



- \* Another way of saying the same thing:
- the number of pi electrons is equal to an odd-numbered pair:
- 1 pair, 3 pairs, 5 pairs, 7 pairs.... (This also produces the series 2, 6, 10, 14 ...)

#### (iv) Condition - 4: The Molecule Must Be Flat

- The fourth condition for aromaticity is that the molecule must be **flat** (**planar**).
- Aromaticity is such a stabilizing property (worth **20-36 kcal/mol**) that generally a molecule that is
  - o cyclic
  - o conjugated
  - o has [4n+2] pi  $(\pi)$  electrons

#### Condition #4: The molecule must be flat

Generally, if the first three conditions are met then it's usually safe to assume that the molecule is flat. .

A prominent exception is the isomer of [10]-annulene below left.



not aromatic

CH<sub>2</sub>

aromatic!

Although it is cyclic, conjugated, and has 10 pi electrons, it is not flat due to repulsions between hydrogens that arise when it is in the flat conformation.

Since it is not flat, it is not aromatic.

However, replacing the hydrogens with bonds to a bridging carbon allows all C-C pi bonds to be in the same plane - aromatic!

**NOTE BY: Annulenes** are monocyclic compounds containing alternating ring double bonds, such as benzene, but of different sizes. According to systematic nomenclature, **Benzene** is a [6] annulene, while **Cyclobutadiene** is [4] annulene, whereas **Cyclooctatetraene** is an [8] annulene.



formally [4]annulene, better known as cyclobutadiene



formally [6]annulene, better known as benzene



formally [8]annulene, better known as cyclooctatetraene

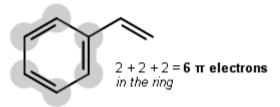
### • Count pi electrons in aromatic compounds

- $\pi$ -bonds are simply the second bond made in a double bond. Any pure double bond is one sigma/ $\sigma$  and one pi/ $\pi$  bond.
- Since any one chemical bond (meaning only one line in bond line notation) contains at most **two electrons**, count two  $\pi$ -electrons per double bond, and ignore the  $\sigma$ -electrons.
- If **lone pairs electron** present, consider the molecular geometry, and only the  $\pi$ -electrons that are in the ring count towards aromaticity.

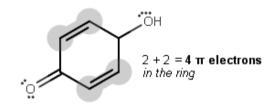


 $2 + 2 + 2 = 6 \pi$  electrons in the ring

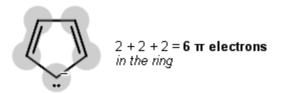
**Aromatic**, because  $4n+2=6\pi$  electrons in the ring (with n=1), planar, fully conjugated all around, and cyclic.



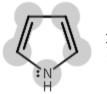
**Aromatic**, because  $4n+2=6\pi$  electrons in the ring (with n=1), planar, fully conjugated all around, and cyclic. The  $\pi$  electrons in the double bond outside of the ring do **not** count towards the  $\pi$  electrons one considers for aromaticity.



**Nonaromatic**, because  $4n+2\neq 4\pi$  electrons, where n must be an integer. It's also not conjugated all around, so it's not antiaromatic. The  $\pi$  electrons in the double bond outside of the ring do **not** count towards the  $\pi$  electrons one considers for aromaticity.



**Aromatic**, because  $4n+2=6\pi$  electrons in the ring (with n=1), planar, fully conjugated all around, and cyclic. The lone pair is actually in a *pure* 2p orbital *perpendicular* to the ring. Don't be fooled, as the alkyl carbon has an implicit hydrogen.



$$2 + 2 + 2 = 6 \pi$$
 electrons in the ring

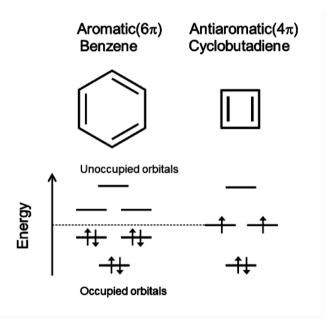
**Aromatic**, because  $4n+2=6\pi$  electrons in the ring (with n=1), planar, fully conjugated all around, and cyclic. The lone pair is actually in a *pure* **2p** orbital *perpendicular* to the ring, which means they count as  $\pi$  electrons.



**Aromatic**, because  $4n+2=6\pi$  electrons in the ring (with n=1), planar, fully conjugated all around, and cyclic. Only one of the lone pairs is actually in a *pure* **2p** orbital *perpendicular* to the ring, which means those counts as  $\pi$  electrons. The other lone pair is actually in a  $\sigma$  (actually,  $sp^2$ ) orbital, so it doesn't count. Thus furan is not **Antiaromatic.** 

# **❖ Huckel's Rule: Aromatic vs. Antiaromaticity**

- Antiaromaticity is a characteristic of a cyclic molecule with a  $\pi$  electron system that has higher energy due to the presence of 4n electrons in it.
- Unlike aromatic compounds, which follow Hückel's rule ([4n+2]  $\pi$  electrons) and are highly stable, **antiaromatic** compounds are highly unstable and highly reactive. To avoid the instability of antiaromaticity, molecules may change shape, becoming non-planar and therefore breaking some of the  $\pi$  interactions.
- The term 'antiaromaticity' was first proposed by *Sir Ronald Breslow* in 1967 as "a situation in which a cyclic delocalisation of electrons is destabilising".
- The criteria for **Antiaromaticity** are as follows:
  - 1) The molecule must be cyclic and completely conjugated
  - 2) The molecule must be planar.
  - 3) The molecule must have a complete conjugated  $\pi$ -electron system within the ring.
  - 4) The molecule must have  $4n \pi$ -electrons where n is any integer within the conjugated  $\pi$ -system [positive integer (n = 0,1,2,3 etc.)].



### • Predicting Aromatic & Anti-aromatic behaviour

- In the first case, the compound must be cyclic, planar (i.e. all the carbon atoms having same state of hybridization) with even number of A value.

$$A = \pi b + e^{-}p + 1$$
(constant)

- where

 $\pi \mathbf{b}$  = number of  $\pi$  bonds with in the ring system

- $\dot{\mathbf{e}}\dot{\mathbf{p}}$  = number of electron pair outside or adjacent to the ring system i.e. if the ring contains hetero atoms (atoms containing lone pair of electrons) which can undergo delocalization and each negative charge if present may be treated as one pair of electrons.
- **Aromatic behaviour:** If the value of 'A', for a certain organic compound comes out as **even number** then this compound will be treated as **aromatic compound**.
- <u>Anti-aromatic behaviour</u>: If the value of 'A', for a certain organic compound comes out as **odd number** then this compound will be treated as **anti-aromatic compound**.
- Order of stability is aromatic > non aromatic > anti aromatic
- Order of reactivity just follows the reverse order of stability as Anti-aromatic > non aromatic > aromatic

Organic Compound (Cyclic, Planar/Cyclic, non-planar)	$\pi$ b value [Number of $\pi$ bonds with in the ring system]	<b>e p value</b> [Number of delocalized electron pair outside or adjacent to the ring system]	A value $[A = \pi b + e^- p + 1]$	Nature of the compound	
Benzene (Cyclic, Planar)	3	0	A = 3+0+1 = 4	Aromatic	
Naphthalene (Cyclic, Planar)	5	0	A = 5 + 0 + 1 = 6	Aromatic	
Anthracene (Cyclic, Planar)	7	0	A = 7 + 0 + 1 = 8	Aromatic	
Cyclopropene (Cyclic, non-planar due to one $sp^3$ hybridized carbon atom)	1	0	A = 1 + 0 + 1 = 2	Non-aromatic	
Cyclopropenyl cation (Cyclic, Planar)	1	0	A = 1 + 0 + 1 = 2	Aromatic	
Cyclopropenyl anion (Cyclic, Planar)	1	1	A = 1 + 1 + 1 = 3	Anti-aromatic	
Cyclobutadiene (Cyclic, Planar)	2	0	A = 2+0+1 = 3	Anti-aromatic	
<b>Cyclopentadiene</b> (Cyclic, non-planar due to one $sp^3$ hybridised carbon atom)	2	0	A = 2+0+1 = 3	Non-aromatic	
Cyclopentadienyl cation (Cyclic, Planar)	2	0	A = 2+0+1 = 3	Anti-aromatic	
Cyclopentadienyl anion (Cyclic, Planar)	2	(For one negative charge on carbon which undergo delocalization)	A = 2+1+1 = 3	Aromatic	
Cyclooctatetraene (Cyclic, Planar)	4	0	A = 4+0+1 = 5	Anti-aromatic	
<b>Cyclooctatrienyl cation</b> (Cyclic, non-planar due to one $sp^3$ hybridized carbon atom adjacent to positive charge)	3	0	A = 3+0+1 = 4	Non-aromatic	
Pyridine (Cyclic, Planar)	3	(Here lone pair on N does not take part in delocalization)	A = 3+0+1 = 4	Aromatic	
Pyrrole	2	1 (Here lone pair on N take part in delocalization)	A = 2+1+1 = 4	Aromatic	
Furan	2	(Here out of two lone pairs on O only one lone pairs take part in delocalization)	A = 2+1+1 = 4	Aromatic	
Cooledesensetes	5	0	A = 5 + 0 + 1 = 6	Not aromatic	
Cyclodecapentaene	Due to the interaction of the hydrogen of 1 and 6 compound become non planar.(combination of steric and angular strain)				
Demons	8	0	A = 8 + 0 + 1 = 9	Aromatic	
Pyrene	Because double bonded $C_{15}$ - $C_{16}$ do not take part in resonance.				

- Which Electrons Count As  $\pi$ -Electrons and which types lone pair contribute to the pi  $(\pi)$  system?
- The total number of pi ( $\pi$ ) electrons for the Cyclopentadiene anion equals 2 (from the lone pair) plus the 4 electrons in the two pi ( $\pi$ ) bonds, giving us a total of 6. This is a Hückel number and the Cyclopentadiene anion is in fact aromatic.

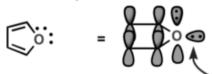
Cyclopentadiene anion: 6 pi electrons

- Here the lone pair can be part of the pi system since the carbon with the lone pair is not already contributing a p orbital to a pi bond.
- Lone pair contributes 2 electrons.
- When added to 4 pi electrons from the 2 pi bonds, we get 6 pi electrons total (a "Huckel number")
- For **pyrrole**, the nitrogen bears a lone pair but is not involved in a pi bond (unlike pyridine, above). Therefore it can contribute to the pi ( $\pi$ ) system and this gives us a total of 6 pi ( $\pi$ ) electrons once we account for the 4 electrons from the two pi ( $\pi$ ) bonds.

Pyrrole: 6 pi electrons

- Similar to cyclopentadiene anion.
- 2 electrons from lone pair contribute to pi system
- When added to 4 pi electrons from the 2 pi bonds, we get 6 pi electrons total (a "Huckel number")
- In **Furan**, the oxygen bears two pairs of lone pair's electron, but it does not means that furan has 8 pi electrons, because each atom can contribute a maximum of one *p*-orbital towards the **pi** ( $\pi$ ) system. In furan, one lone pair is in a *p* orbital, contributing to the **pi** ( $\pi$ ) system; the other is in the plane of the ring. This gives us a total of 6 **pi** ( $\pi$ ) electrons. Furan is aromatic. (So is thiophene, the sulfur analog of furan).

Furan: 6 pi electrons



- Here the oxygen has 2 lone pairs, but only one can be part of the pi system
- When added to 4 pi electrons from the 2 pi bonds, we get 6 pi electrons total (a "Huckel number")

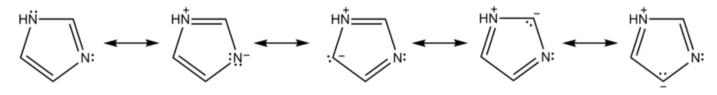
Lone pair at 90 degrees to pi system (don't count as "pi electrons")

- **Imidazole**, which has two nitrogen's. One nitrogen (the N-H) is not involved in a pi bond, and thus can contribute a full lone pair; the other is involved in a pi bond, and the lone pair is in the plane of the ring. This also gives us a total of  $6 \text{ pi } (\pi) \text{ electrons}$  once we account for the **two pi**  $(\pi) \text{ bonds}$ .

Imidazole: 6 pi electrons

 Only the nitrogen which is not part of a pi bond can donate a lone pair

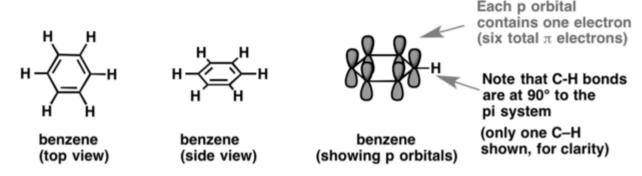
Lone pair at 90 degrees to pi system (don't count as "pi electrons")



#### - Pyridine and the Benzene Anion

- o In benzene, each *p*-orbital is arranged at right angles (90°) to the plane of the ring. Each *p*-orbital contains a single electron.
- ο The total number of **pi** ( $\pi$ ) **electrons** in **Benzene** by counting the pi bonds: **3 pi** ( $\pi$ ) **bonds** times two electrons = **6 pi** ( $\pi$ ) **electrons** total.

### p Orbitals Contribute To Aromaticity in Benzene



- o The **Benzene Anion** has a **lone pair** on one of the carbons. This lone pair can't be in a *p*-orbital, since the *p*-orbital is participating in the **pi** ( $\pi$ ) system. Instead, it's at 90 degrees to the **pi** ( $\pi$ ) system, in the plane of the ring.
- o This lone pair electron on carbon doesn't count as a pair of pi ( $\pi$ ) electrons since it can't overlap with the pi ( $\pi$ ) system.
- o Pyridine, where the lone pair is also at right angles to the pi  $(\pi)$  system, but it is participating in the pi  $(\pi)$  system.

### Lone pairs in the "Benzene anion" and Pyridine are not actually "Pi electrons"

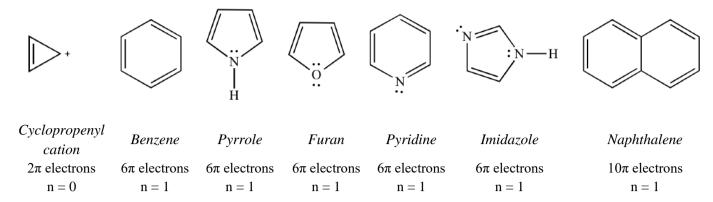
In the "benzene anion", the carbon with the lone pair is already contributing a p orbital (and one pi electron) to the pi system, just as in benzene.

The lone pair is at 90° to the pi system (in the "plane of the ring")
Since it can't overlap, its two electrons therefore don't count as "pi electrons"

Don't count as "pi electrons"

The same is true for pyridine

#### **Aromatic Molecules**



Summary: Anti-Aromaticity						
Aromatic	Anti-Aromatic	Non-Aromatic				
unusually stable	unusually unstable	everything else				
·Cyclic	•Cyclic	Fails any one of the 4 criteria on the left				
·Conjugated	·Conjugated					
·(4n+2) Pi electrons	·(4n) Pi electrons					
•Flat	•Flat					
e.g.	e.g.	e.g.				
benzene	cyclobutadiene	cyclooctatetraene				
(resonance energy of 36 kcal/mol)	(only stable below -100 °C )	(adopts a tub-like shape to "escape" anti-aromaticity)				

- Aromatic molecules are cyclic, conjugated, have (4n+2) pi  $(\pi)$  electrons, and are flat.
- Anti-aromatic molecules are cyclic, conjugated, have (4n) pi ( $\pi$ ) electrons, and are flat.
- Non-aromatic molecules are every other molecule that fails one of these conditions.

### • What is the major difference between an antiaromatic and aromatic compound?

- (a) The structure must be cyclic for aromatic but not antiaromatic compounds.
- (b) Antiaromatic compounds have at least one  $sp^3$  hybridized atom in the ring
- (c) Antiaromatic compound can assume a chair-like structure while aromatic compound are nearly flat
- (d) Aromatic compounds cannot have a charged atom in the structure
- (e) Only aromatic compounds follow **Huckle's rule**.

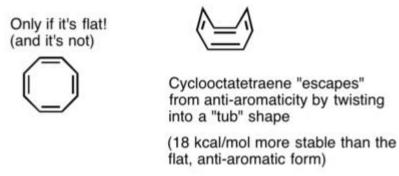
#### • Why antiaromatic compounds are highly unstable?

- Greater the delocalisation energy (resonance energy) of a compound, more stable it is. The resonance energy can be calculated using **Huckel Molecular Orbital Theory** (HMOT). Antiaromatic compounds have **zero resonance energy hence are unstable**.
- According to HMOT,

Delocalisation energy=( Pi electron binding energy) - (total energy of isolated double bonds in a classical strucure)

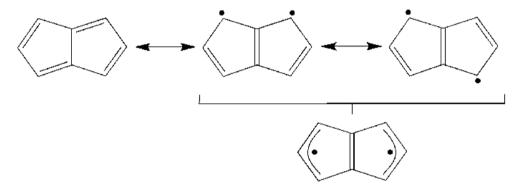
### • Why Cyclooctatetraene "Escapes" Anti-Aromaticity

- Cyclooctatetraene is anti-aromatic only if it is flat. However, the relatively "floppy" structure of Cyclooctatetraene allows for some flexibility. The bonds can rotate away from flatness such that the molecule adopts a "tub-like" shape, thereby avoiding the "Antiaromaticity tax" of 18 kcal/mol that would be paid if all the *p*-orbitals on the molecule were conjugated with each other.



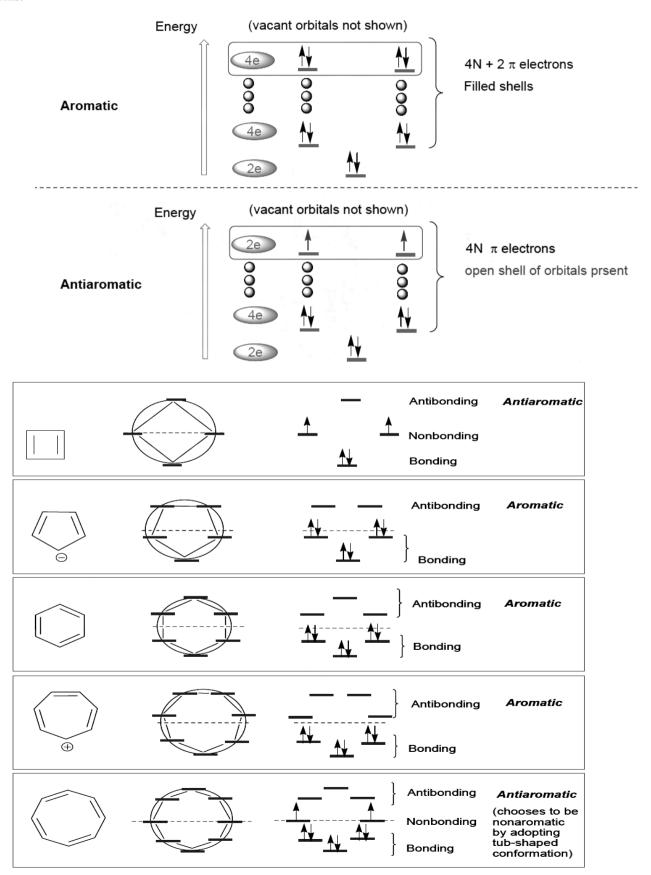
### • Why Pentalene stuck in its anti-aromatic conformation

- Pentalene (above) which also has **8 pi** ( $\pi$ ) electrons has a very rigid bicyclic structure that prevents bond-rotation away from flatness. Hence, it's stuck in its anti-aromatic conformation.



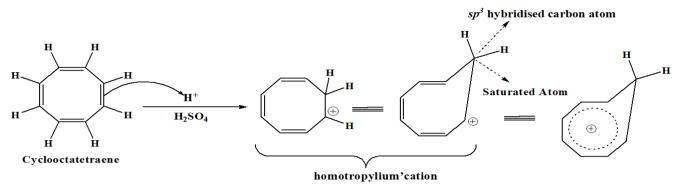
### • Points to remember while making predictions on aromaticity using Frost's circle

- Aromatic compounds will have all occupied molecular orbitals completely filled whereas antiaromatic compounds would have incompletely filled orbitals.
- If an antiaromatic system (4n electrons) has the freedom to undergo conformational change and become nonaromatic that would do so. Remember that antiaromatic state is less stable than aromatic and nonaromatic forms.



# **\*** Homoaromaticity

- **Homoaromaticity**, in organic chemistry, refers to a special case of aromaticity in which conjugation is interrupted by a single  $sp^3$  hybridised carbon atom.
- That means if a stabilized cyclic conjugated system (4n+2 e s) can be formed by bypassing one saturated atom, that lead to *Homoaromaticity*.
- The concept of Homoaromaticity was pioneered by Sir Saul Winstein in 1959.



If a satisfied cyclic conjugated system  $[(4n+2)\pi e]$  can be formed by passing one saturated atom that will lead to the so called Homoaromatic compound.



*Sir Erich Huckel* [Erich Armand Arthur Joseph Huckel] was a German physicist; physical chemist was born on August 09, 1896 – died on February 16, 1980. Huckel is famous for his greatest contributions towards Debye-Huckel theory and Huckel method.

In the year of 1923 along with Debye developed their theory of electrolytic solutions, elucidating the behaviour of strong electrolytes by considering interionic forces, in order to account for their electrical conductivity and their thermodynamic activity coefficients.

Hückel is most famous for developing simplified quantum mechanics methods to deal with planar unsaturated organic molecules.



Organic chemist *Sir Ronald Breslow*, Born on March 14, 1931- Died October 25, 2017 (aged 86) New York, New York, U.S.

Professor **Breslow's** research focuses on the design and synthesis of new molecules with interesting properties. His interests include anti-aromaticity, biochemical reactions, remote functionalization reactions, the development of artificial enzymes, and cytodifferentiating agents. He has been involved in the synthesis of the cyclopropenyl cation, as well as in the development of the histone deacetylase inhibitor SAHA (Vorinostat) which is used for the treatment of lymphoma.

**Awards:** ACS Award in Pure Chemistry (1966); NAS Award in Chemical Sciences (1989); National Medal of Science (1991); Priestley Medal (1999); Othmer Gold Medal (2006); Perkin Medal (2010); AIC Gold Medal (2014).



Sir Saul Winstein was the Canadian chemist who discovered the Winstein reaction, in which he argued a non-classical cation was needed to explain the stability of the norbornyl cation. He also first proposed the concept of an intimate ion pair. He attended the University of California, Los Angeles, where he completed his work for an B. A. degree in 1934, and a M.A. degree in 1935. In 1938 he received the Ph.D. degree from the California Institute of Technology. After a postdoctoral fellowship at Caltech, he spent 1939-40 as a National Research Fellow at Harvard University associated with Professor Paul Bartlett. After a year as Instructor at the Illinois Institute of Technology in 1940-41, he returned to his alma mater, UCLA, which he served as Instructor (1941-42), Assistant Professor (1942-45), Associate Professor (1945-47), and Professor (1947 to 1969). He received the National Medal Of Science in 1970. He died on November 23, 1969 in Los Angeles, California, United States.