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Single Covalent Bonding Structure in Fullerenes, Carbon Nanotubes and Closed Nanotubes

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Abstract

The present paper deals with carbon in highly organized solids like graphene and its three-dimensional derivatives: fullerenes, carbon nanotubes and capped carbon nanotubes. It proposes an alternative to the typical bonding pattern exposed in literature. This novel bonding pattern involves alternating positively and negatively charged carbon atoms around hexagonal rings, then a few uncharged and partially bonded atoms close to the pentagon rings. The article focuses on fullerenes inscribed into a regular icosahedron, then addressing the most common fullerenes like C60. Carbon atoms are found to have predominantly three single bonds and less often two separated single bonds. The same pattern explains equally well carbon nanotubes and closed-tip nanotubes, of which C70 is a special case.

Keywords

Fullerenes, Icosahedron, Carbon Nanotube, Chemical Bond, Even-Odd Rule, Graphene, C60, C80, Triple Series

1. Introduction

Before the discovery of fullerenes, the known allotropes of carbon were diamond and graphite. Both have crystalline structures, in which carbon atoms are neutral atoms with a tetravalent bonding structure [1]. In 1970, E. Osawa predicted the existence of a sphere formed with 60 carbons, the first fullerene [2]. These were later experimentally produced in complex conditions involving vaporizing graphite with laser irradiation [3]. Fullerenes are composed of carbon atoms forming nearly spherical molecules, with a solid surface around an empty volume [4]

[5] [6]. This surface has nearly the same bonding structure as graphene [7] resembling a rolled sheet of graphene. Numerous characterizations where performed to understand the exact arrangement of carbons, bring to light hexagonal rings of carbon with a few pentagon rings [8] [9] [10] [11] [12]. In graphene, fullerenes, and carbon nanotubes alike, each carbon atom has three neighbors. The generally accepted bonding rules impose two single bonds and one double bond [13] [14].

In previous work, G. Auvert has proposed that only electrically charged carbon atoms can erect three bonds, whereas neutral carbon atoms may only have 2 or 4 bonds [15] [16] [17]. The present paper focuses on the consequences of carbon bonding capacities on the bonding pattern in fullerenes, carbon nanotubes and closed-end carbon nanotubes. It will be considered that the most stable configurations imply that most carbons bear an electrical charge with three bonds and sometimes the possibility that some bonds are broken leaving some neutral carbons with two single bonds.

For the sake of example, the authors have chosen to focus on fullerenes having a regular repartition of pentagons, using the Euler characteristic [4] [18]. The present article hence starts by describing the geometrical repartition of carbon atoms in fullerenes that can be inscribed in an icosahedron. Then, connections rules between neutral and charged carbons are applied, leading to bonding patterns and their two or three-dimensional representations. A focus is later made onto carbon nanotubes and caped nanotubes. In the discussion, classical and recent connection rules are compared, as well as implication on endohedral species. The special case of C20 is also addressed.

In this paper, C20 or C120 represent molecules of fullerenes with carbon atoms.

Note: To draw fullerenes and nanotubes, two software are used [19] [20].

2. Locating Carbon Atoms of Fullerenes over an Icosahedron

Graphene, fullerene, and carbon nanotubes are composed of equidistant carbon atoms, measured to be between 0.134 and 0.154 nm [14] [21]. Moreover, in graphene layers and in nanotubes, carbons form regular hexagonal structures [13]. In spherical fullerenes however, carbons form two different types of rings: pentagonal and hexagonal rings [5]. We know from Euler's theorem that only twelve pentagons are always present in fullerenes (*the* 12 *Pentagon Theorem*) [4] [5] [6] [18]. To obtain a regular repartition of these twelve pentagons, carbon atoms can be positioned on the facets of a virtual convex regular icosahedron [22]. As illustrated in **Figure 1**, this specific icosahedron has twelve vertices forming twenty equilateral triangles.

To maintain the regularity of the atom structure, carbon atoms must be regularly disposed over the twenty triangular facets.

In scientific literature, C60 is known to be a truncated icosahedron [3].

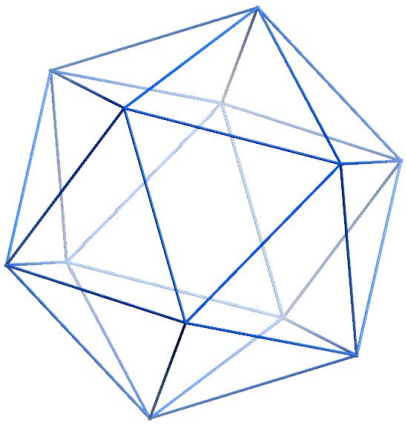

Figure 1. Drawing of a convex regular icosahedron with twenty equilateral triangle, thirty edges and twelve vertices. All fullerenes studied here have their carbon atoms localized on the surface of this icosahedron.

2.1. The Icosahedron Model Can Be Scaled Up

A huge variety of carbon positions is possible over the facets of the icosahedron. To maintain equidistance between carbon atoms however, the most obvious repartition involves placing some of the atoms on the edges and the other regularly over the facets. With this constrain in mind, we can derive a series of fullerenes by multiple of three, and by increasing the number of atoms on facets. Table 1 lists fullerenes belonging to what will be named the "Triple-series".

Table 1. Number of carbons in fullerenes belonging to the "Triple-series". In this series, n is a positive integer. The number written after the letter C gives the number of carbons in the fullerenes. The icosahedron has twenty facets. Three is the number of carbons in the smallest triangle.

Number n	Triple-series
n	$C(20 * 3 * n^2)$
n = 1	C60
n = 2	C240
n = 3	C540
n = 4	C960
n = 5	C1500

This series can be expressed with $C(20 * 3 * n^2)$, where n is a positive integer of unlimited value and the resulting figure in bracket the number of carbon atoms in a regular icosahedron forming fullerenes. The smallest fullerene in the Triple-series is C60, the most observed regular fullerene. C60 has three atoms per triangles, hence the name of this family.

Icosahedrons facets are equilateral triangles on which several carbon atoms are located. To clarify the position of the atoms and keep equidistance, these triangles will be sub-divided into elementary equilateral triangles.

2.2. Three Carbons in Each Elementary Triangle

In the following, each of the 20 triangles of the icosahedron is divided in (n^2) equilateral triangles named "elementary triangle".

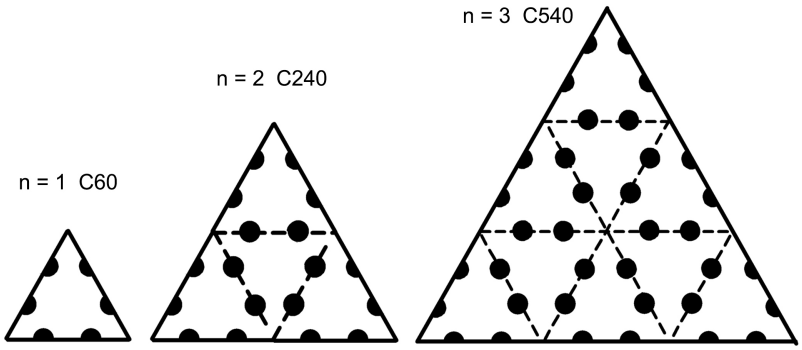

Figure 2. Atom repartition over one facet of the icosahedron for the first three fullerenes belonging to the "Triple-series": C60, C240 and C540. Black dots represent carbon atoms which are shared over elementary triangles. They also clearly form regular hexagons all over facets of the icosahedron. The equidistance between atoms is satisfied and conforms to what is experimentally observed in fullerenes.

As shown in **Figure 2**, six carbon atoms are distributed on the edges of each elementary triangle. Since these atoms are shared between contiguous elementary triangles, the number of carbon atoms on an elementary triangle is three, and the total number of atoms is three times that of the number of elementary triangles.

2.3. Two-Dimensional Drawing of Fullerenes

Coming back to the global view of fullerenes, **Figure 3** illustrates a flat view of C60 (n = 1, the smallest fullerene in the "Triple-Series"). It gives a better view of the relative positions of carbons forming pentagons and hexagons.

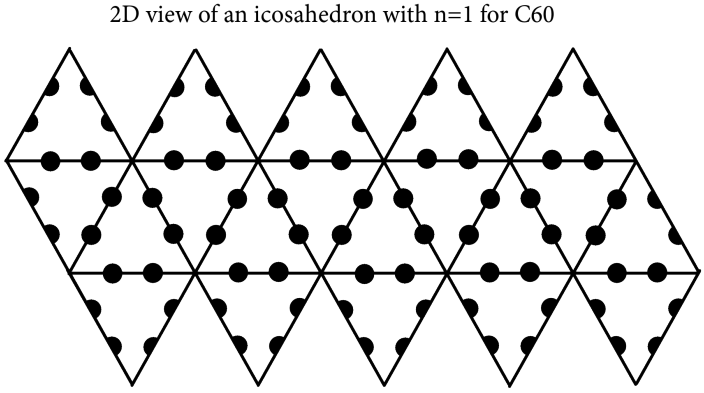

Figure 3. Flat view of C60 fullerene with carbon atoms located on the edges of the icosahedron facets. Carbon atoms form regular pentagonal rings around each vertex and hexagonal rings inside each facet. This construction ensures that the distance between neighbor atoms is always the same, as experimentally observed in fullerenes. In this view, twelve vertices are placed where five triangles meet.

In **Figure 3**, C60 has 20 hexagons [4] (p 280), one hexagon in each of the 20 triangles, or facets of the icosahedron. All twelve vertices of the icosahedron overlook five carbon atoms forming twelve pentagon rings (see also in the first chapter of the discussion for a three-dimensional drawing).

3. Bonding Conditions between Carbons in Fullerenes

IUPAC define the suffix -ene as designing structures containing carbons atoms that are bonded with three atoms, over four covalent bonds that are not fully distinct [23] [24]. This is usually represented as two single bonds and one double bond. With three neighbors to each carbon atom, fullerenes belong to the -ene group and the classical bonding structure prevails in literature as shown for C60 in [3] [4] [25].

In this chapter, the authors use an alternative rule previously published under the name "even-odd" rule [15]. Applied to carbon atoms, it implies that:

- two neighboring carbon atoms can only be connected through a single covalent bond,
- carbon atoms bearing a charge can have only three covalent bonds,
- neutral carbon atoms on the contrary can only erect two or four distinct covalent bonds. In fullerenes, because carbon atoms are organized in planes (hollow molecules) neutral carbon atoms have two bonds,
- connected charged carbons bear opposite charges.

In fullerenes, carbons atoms are mainly organized in hexagonal rings, each with three neighbors. Following the above rule, each hexagon is an alternating pattern of positively and negatively charged atoms, with each atom bonded to their three neighbors. This alternating pattern is interrupted by the presence of pentagons, solved by a neutral carbon atom with two bonds only. Each neutral carbon atoms faces another neutral atom, resulting in a void, a missing bond. The alternance of charges ensures the overall neutrality of the fullerene compound. For a better understanding, **Figure 4** illustrates the "Triple-series" fullerene bonding pattern with the C60, the smallest fullerene of the "Triple-series".

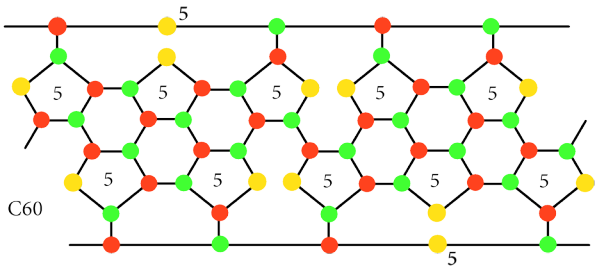

Figure 4. C60 fullerene with bonded and unbonded carbons as a two-dimensional map drawn with *ChemSketch* [19]. Red and green carbons bear opposing charges and have three bonds. In each pentagon, highlighted by the label "5", one neutral (yellow) carbon atom has two bonds. There are 12 neutral carbons forming six pairs. The ratio between yellow carbons versus all carbons is 20%. Details of the drawing procedure in the first chapter of the discussion. C60 is a neutral compound; this is ensured by the presence of 24 positive carbons, neutralized by 24 negative carbons.

C60 in Figure 4 is very stable, thanks to the uniform charge repartition and the bonding mesh in hexagons. Said differently, a continuous path with alternating positively and negatively charged atoms can be followed without interruption all around the circumference of the fullerene.

Figure 5 shows three-dimensional representations of the three smallest fullerenes in the Triple-series. Uncharged carbons have two bonds and form facing pairs along some of the edges of the icosahedron.

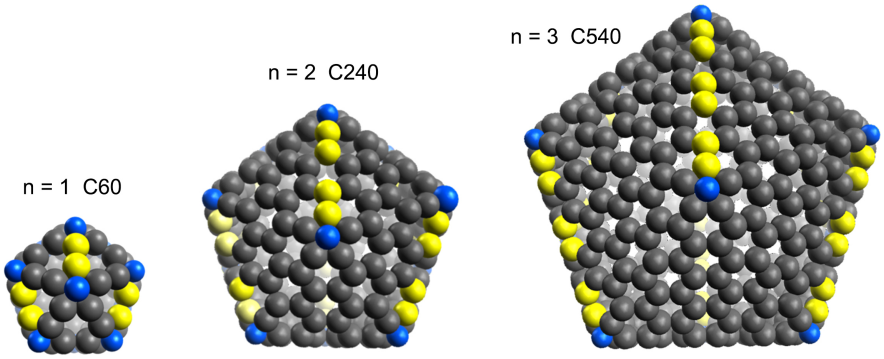

Figure 5. Three-dimensional top-view of the three smallest fullerenes in the Triple-series. Black spheres represent charged carbon atoms with three bonds and yellow spheres are neutral carbon atoms with two bonds. Blue spheres mark the icosahedron vertices, located above the centers of the pentagons (*they are not atoms*). The hexagonal pattern can be observed everywhere else. Triangular facets of the icosahedrons are delimited by three blue spheres. In C540, the biggest drawing, six neutral yellow carbons are aligned on some edges, between two pentagons. They form a chain between pentagon pairs, showing that only six groups of carbon pairs can exist in the studied fullerenes.

It is possible to calculate the number of unbonded carbon atoms in fullerenes as a multiple of n: 6 * n. In Table 2, the number of unbonded carbon pairs, *i.e.* pair of uncharged atoms, in the first fullerenes of the "Triple-series" is reported. Uncharged carbon atoms thus represent (20/n)% of the overall number of atoms in a fullerene of the "Triple series". This decreasing factor favors stability of the compound even for large values of n.

Table 2. Number of uncharged carbons pairs without bond in the Triple-series. The first line allows to calculate these numbers in which n is a positive integer.

Number n	Triple-series	Unbonded Carbon Pairs
n	C($20 * 3 * n^2$)	6 * n
1	C60	6
2	C240	12
3	C540	18
4	C960	24
5	C1500	30

4. Carbon Nanotubes Closed with Fullerenes

We will now turn our attention to the bonding pattern of nanotubes and caped nanotube.

Nanotubes have been intensively studied since 1952 [26] [27] [28]. They are empty cylinders [29] with an external surface composed of carbon atoms organized like in graphene [13]. Ends of nanotubes have been experimentally observed, with evidence that nanotubes are sometimes caped [30] [31]. We will explain how semi-hemispheric fullerenes can close a nanotube of the same width and how the pattern observed earlier in fullerenes is still applicable. The usual bonding structure involves two single-bonds and one double-bond [30].

Let us concentrate on nanotubes for which the total number of atoms can be expressed as C(10 * n * m), where m is the number of layers, 10 * n the number of carbon atoms in one layer and nthe same number as used above in fullerenes. Views from the tip and side of a tube with ten-atom transverse layers (n = 1) and eight planes (m = 8) are drawn in Figure 6.

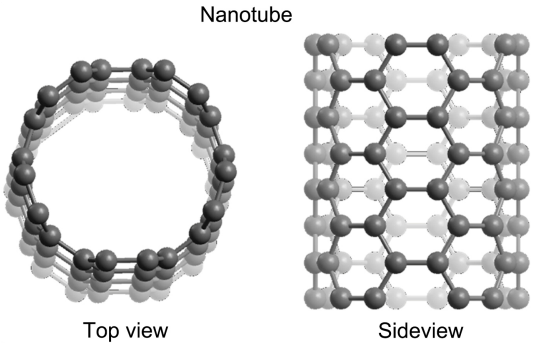

Figure 6. View from the tip and side view of open-ended carbon nanotubes with chiral indices (5, 5) with 10 carbons in each layer [29]. The hexagon pattern is the same than in fullerenes and graphene.

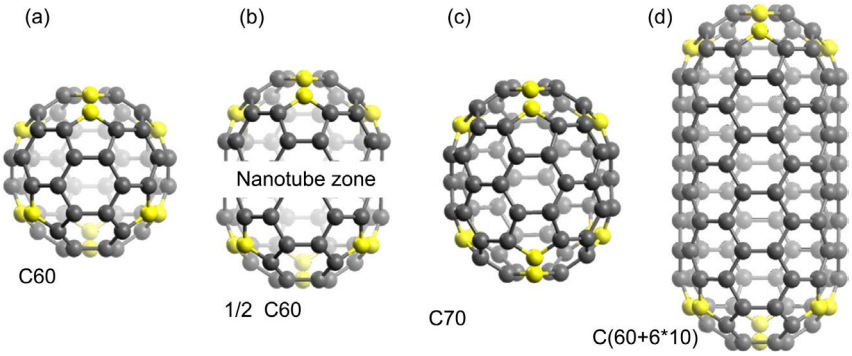

Figure 7. Composition of a single-wall carbon nanotube closed with half C60 (n = 1). (a) C60 fullerenes with uncharged atoms highlighted in yellow. (b) two halves of a C60 (c) C70, composed of two half-C60 and a layer of 10 carbon atoms (d) longer nanotube of width 10; chiral indices (5, 5); with 6 levels. Each cap can be rotated several times with a 72˚ angle without changing electrical configurations of this closed-tip nanotube. If the number of layers in the tube becomes uneven, one of the closing half fullerenes will rotate by 36˚.

In his 1987 article, H. W. Kroto has pointed out that C70, the other most stable fullerene, is formed by separating two halves of a C60 by a ring of ten extra carbon atoms [25]. These extra atoms can be likened to a short nanotube. Extending the rationale, the number of layers could be of any size, forming longer nanotubes (C80 with m = 2, C100 with m = 4 …). In other words, nanotubes of chiral indices (5, 5) are naturally caped by half C60. Having observed this, the bonding pattern described in the above paragraphs can be used to close nanotubes, as illustrated in Figure 7. The body of the nanotube is composed of alternating positively and negatively charged carbons, connected with three bonds. At each end, six neutral carbon atoms are bonded only twice.

5. Discussions

Discussions focus on four topics. First why a neutral atom is the only solution for carbon pentagon rings; then a quick note on the structure of C20 fullerenes, which doesn't belong to the "Triple-series"; next a thought on endohedral species and last with a comparison of different representations of the C540 fullerenes.

5.1. Drawing Charges Positions in Hexagons and Pentagons

To build flat-views of fullerenes as in Figure 3 and Figure 4, carbons are positioned at equidistance. Then, all the connections are drawn in one procedure in which carbons with four bonds are not allowed. Starting on one carbon atom with three bonds, its charge is set to be positive for instance. A path is chosen, and alternating charges are written until the path loops back to the starting carbon. If the alternance is maintained all the way, this way is electrically acceptable (the number of carbons in the path is even). Otherwise, some bonds must be modified, and neutral atoms appear. The above procedure is illustrated in Figure 8. For each member of the "Triple-series" described in this paper, there is at least one possible path (see Figure 4).

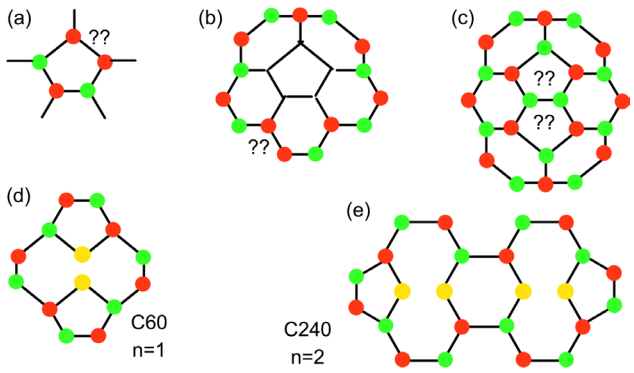

Figure 8. Planar view of carbons atoms with two or three covalent bonds. Different attempts at bonding patterns are shown. Carbons in green color are positively charged. In red color, they are negatively charged. To be valid a path between carbon must never involve two successive carbons bearing the same charge. (a) around a pentagon, the path does not loop back properly (odd number of atoms). (b) and (c) also are not possible. (d) and (e) paths are possible when adding pairs of neutral carbons (in yellow).

5.2. Stability of the Smallest Fullerenes C20

The C20 fullerene is an interesting case although it does not belong to the "Triple-series" described earlier in this paper. C20 can also be inscribed in an icosahedron, but carbon atoms are placed this time in the center of each facet. C20 is composed of 12 pentagons and no hexagons. Classical bonding rules impose two single bonds and one double bonds for each carbon. This bonding structure, as illustrated in Figure 9, seems very homogenous and therefore very stable.

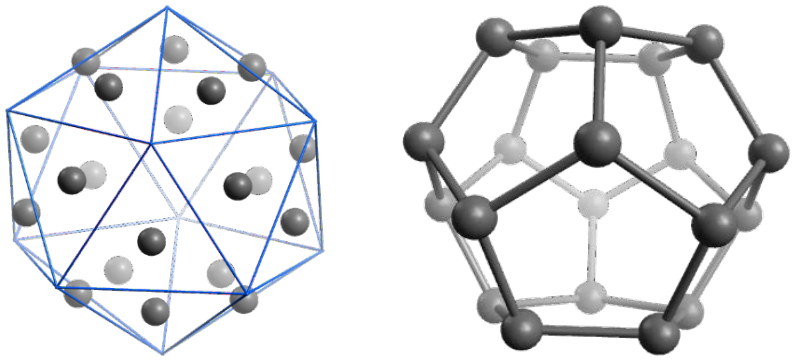

Figure 9. Left: three-dimensional view of a C20 inscribed in an icosahedron. Blue lines in these drawings represent a convex regular icosahedron with twenty triangular facets [22]. Twenty carbon atoms are placed in the center of the facets. Right: View of the C20 without icosahedron edges. In the classical theory, each carbon atom is bonded to its three neighbors.

With the even-odd rule however, the structure involves twelve neutral carbon atoms, which represents 60% of the overall number of atoms in this compound. Both bonding patterns are illustrated in Figure 10, where the classical bonding pattern available in [4] can be compared to the structure obtained with the even-odd rule. The many missing bonds point toward high instability of this compound. Another comparison can be made with the stable graphene structure, composed of charged carbons with three bonds in a single flat layer.

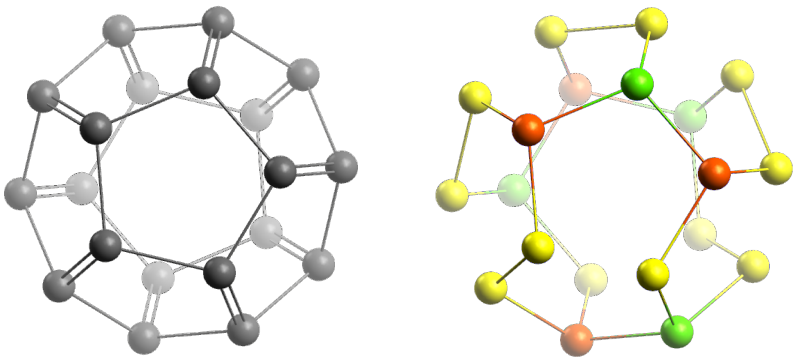

Figure 10. Left: Three-dimensional view of C20 with classical bonding configuration. Right: C20 bonding structure derived with the even-odd rule. This structure seems less stable *i.e.* not enough bonds.

These drawings would at first indicate that the classical bonding structure better explains C20, if C20 had indeed been experimentally observed. As a matter of fact, only stabilized versions of C20 have been observed or synthesized. In his 1987 article, W. H. Kroto stated that molecules with fused pentagons, i.e. pentagons sharing at least an edge, are unstable [25]. The highest stability is reached once each pentagon is surrounded with five hexagons. The present article can be taken as a demonstration of Kroto's rule for C(<60).

Let us take a closer look at a stabilized version of the C20, C20H20, produced and characterized [32]. In this case, the hydrogen atoms create a tetravalent bonding structure around each carbon atoms, allowing for uncharged atoms and four bonds, as in diamond. The resulting compound is much more stable.

The "Triple-series" described earlier, on the other hand, is first composed of the most observed regular fullerenes [4] and second follows Kroto's rule.

5.3. Endohedral-Fullerenes

Endohedral-fullerenes species are fullerenes that have additional atoms enclosed within their inner spheres [33].

The first ever observed were endohedral metallofullerenes La@C60 in 1985 [34]. Enclosed atoms can be noble gases, metal or other molecules that do not bond with the carbon atoms of the fullerene cage. Incorporation of these additional atoms requires specific conditions like high pressure or high temperature (see the example of Xenon in [35]). In a previous work, Saunders *et al.* have assumed that a temporary window opens under specific conditions [36]. Our hypothesis is that voids near the pentagon rings of the fullerene are actual permanent windows through which neutral atoms penetrate inside the cage.

5.4. Three-Dimensional Representations of Large Fullerenes

There are very few three-dimensional representations of large fullerenes to be found. We offer a physical representation of C540 in **Figure 11**, complying with the "even-odd" rule. For the sake of comparison, we searched for alternative representations shown in **Figure 12**.

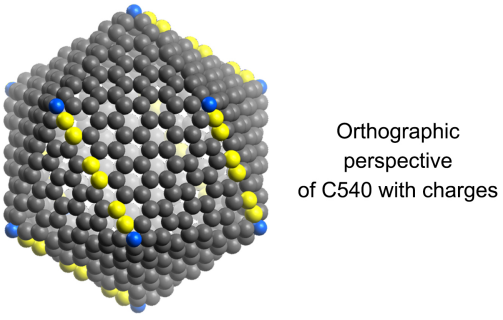

Figure 11. Orthographic perspective of C540 of the Triple-series. Yellow-colored atoms are uncharged and bonded twice while black-colored atoms are charged and bonded thrice. The blue points symbolize vertices and are above pentagons rings. Triangular facets of the enclosing icosahedron are clearly seen.

In available drawings, double or single bonds are not represented as such, but every carbon atom has three bonds. These representations are usually more mathematical or computational views than physical likenesses.

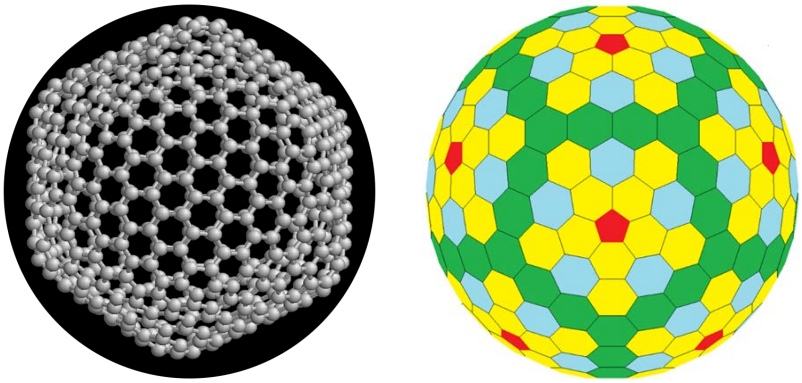

Figure 12. Commonly found representations of C540 with all carbon atoms having three bonds. Left: from Brian0918 [5]. Pentagon rings are in the same location than in **Figure 11**. Right: sphere-like representation [37], without classical double bonds and heterogenous C-C distances. In this drawing, pentagons' positions are a little different than in our drawing. In both representations, no specific charges or double bonds are represented.

6. Conclusion

The present article has proposed a bonding pattern for regular fullerenes like C60 and bigger fullerenes of type C(20 * 3 * n^2), obtained by applying the recently published even-odd rule. As an alternative to the classical model, the pattern involves alternating positively and negatively charged carbon atoms, then a few uncharged and partially bonded atoms close to the pentagon rings. First, we have modelled C60 and other regular fullerenes as being inscribed in an icosahedron form. Then, we derived a family of fullerenes belonging to a specific geometrical pattern that preserves equidistance between carbon atoms, which we named the Triple-Series, of which C60 is the smallest. The same bonding pattern applies to all fullerenes of this family, satisfying both overall compound neutrality and local stability, with a maximum of 20% of atoms partially bonded. Next, we used the notion that nanotubes can be capped with half-fullerenes of the Triple-Series to derive their bonding pattern. C70 is a special case of this sort and the bonding pattern points toward good stability. Each elongation by ten carbon atoms is also stable. In contrast, the same bonding rules were applied to C20, not in the Triple-Series, and the resulting pattern displays many missing bonds, indicating compound instability. We conclude that the even-odd rule can be used to explain why C60 and its derivative are the most common fullerenes and why the C20 is unstable unless stabilized.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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