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Fullerene Graphs and Some Relevant Graph Invariants

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1 Introduction

Fullerenes are polyhedral molecules made entirely of carbon atoms. They come in a wide variety of sizes and shapes. The most symmetric is the famous buckminsterfullerene, \( C_{60} \), whose discovery in 1985 marked the birth of fullerene chemistry \cite{40}. The name was a homage to Richard Buckminster Fuller, whose geodetic dome it resembles. Since 1991, when the Science magazine pronounced the buckminsterfullerene for the “Molecule of the year”, fullerenes have been attracting attention of diverse research communities. The experimental work was paralleled by theoretical investigations, applying the methods of graph theory to the mathematical models of fullerene molecules called fullerene graphs. One of the main driving forces behind that work has

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been a desire to identify structural properties characteristic for stable and reactive fullerenes, i.e., for fullerene isomers verified in macroscopic quantities.

Ante Graovac also found fullerenes as an interesting topic to study [27, 36, 44], he connected the well known topological indices with the fullerene structures [23, 30].

1.1 Fullerene graphs

Fullerenes can also be seen as graphs, vertices represent atoms, and edges represent bonds between atoms. A fullerene graph is a 3-connected 3-regular planar graph with only pentagonal and hexagonal faces. By Euler’s formula, it follows that the number of pentagonal faces is always twelve. Grünbaum and Motzkin [34] showed that fullerene graphs with \( n \) vertices exist for all even \( n \geq 24 \) and for \( n = 20 \). Although the number of pentagonal faces is negligible compared to the number of hexagonal faces, their layout is crucial for the shape of a fullerene graph. There are fullerene graphs where no two pentagons are adjacent, i.e., each pentagon is surrounded by five hexagons. Those fullerene graphs satisfy the isolated pentagon rule or shortly IPR, and they are the most stable fullerene compounds [41]. If all pentagonal faces are equally distributed, we obtain fullerene graphs of icosahedral symmetry, whose smallest representative is the dodecahedron. The dodecahedron is the only icosahedral fullerene that does not satisfy the IPR. On the other hand, if the pentagonal faces are grouped in two clusters by six, we obtain nanotubical fullerene graphs.

1.1.1 Icosahedral fullerene graphs

The common feature of all icosahedral fullerenes is their geometrical shape. The simplest icosahedral fullerene graph is the dodecahedron, \( C_{20} \). Caspar and Klug [6] and Coxeter [7] suggested a method that works with the icosahedral fullerene graph dual: geodesic domes, i.e., triangulations of the sphere with vertices of degree 5 and 6.

Goldberg [29] observed that all icosahedral fullerene graphs can be obtained by mapping (a part of) the hexagonal grid onto the triangular faces of an icosahedron. He also showed that the number of vertices \( n \) in a polyhedron of icosahedral symmetry can be determined by two integers \( i \) and \( j \) by the following equation, conveniently
called the Goldberg equation

\[ n = 20(i^2 + ij + j^2). \]  

(1)

The integers \( i \) and \( j \) in the Goldberg equation are in fact the components of a two-dimensional Goldberg vector \( \vec{G} = (i, j) \), sometimes also called Coxeter coordinates. To avoid the mirror effect, we always assume that \( 0 \leq i \leq j \) and \( 0 < j \).

![Figure 1: Construction of a (2,3)-triangle, a metaface of a (2,3)-icosahedral fullerene graph. The vertices of the equilateral triangle ABC are centers of pentagons.](image)

This vector determines the distance and positions of the vertices of the \( (i, j) \)-triangle in the hexagonal lattice. See Figure 1 for a construction method of an \( (2, 3) \)-triangle. Precisely 20 such \( (i, j) \)-triangles produce an \( (i, j) \)-icosahedral fullerene in a manner shown on Figure 2. The vertices of the triangles are centers of the 12 pentagons of the fullerene graph. The pair of the triangles \( ABC \) and \( EB'C' \), as shown on Figure 2, is a pair of opposite triangles. There are exactly 10 such pairs. The pentagons with centers in \( A \) and \( E \) (Figure 2) are called antipodal pentagons (similarly, the pentagons with centers in \( B \) and \( B' \), as well as those with centers \( C \) and \( C' \) are antipodal).

The icosahedral group \( I \) is the group of rotational symmetries of the icosahedron and dodecahedron, of order 60. This group is isomorphic to \( A_5 \), the alternating group of even permutations of five objects. The full icosahedral group \( I_h \), is the point group of all symmetries of the icosahedron and dodecahedron. This group is equivalent to the group direct product \( A_5 \times \mathbb{Z}_2 \) of the alternating group \( A_5 \) and cyclic group \( \mathbb{Z}_2 \). The order of the full icosahedral group of symmetries is 120. The \( (i, i) \)- and \( (0, i) \)-
icosahedral fullerene graphs, $i > 0$, have full icosahedral symmetry group, i.e., every element of this class of graphs has a symmetry group $I_h$.

The idea of Goldberg was then used by Coxeter [7] and Graver [31] to classify highly symmetric fullerene graphs.

1.1.2 Nanotubical fullerene graphs

While the icosahedral fullerenes have “spherical” shape, there is a class of fullerene graphs of tubular shapes, called nanotubical graphs or simply nanotubes. They are cylindrical in shape, with the two ends capped with a subgraph containing six pentagons and possibly some hexagons. The cylindrical part of the nanotube can be obtained by rolling a planar hexagonal grid. The way the grid is wrapped is represented by a Goldberg vector $(i, j)$, called also the type of the nanotube. See Figure 3 for an example of the construction of the cylindrical part of a nanotube.

Nanotube fullerene graphs exist for all vectors $(i, j)$ with $i + j \geq 6$ and for the vector $(0, 5)$. A $(0, 5)$ nanotube is depicted on Figure 5.

Nanotubes with $i = 0$ are called zig-zag nanotubes, and the ones with $i = j$ are called armchair nanotubes. These are the only types of nanotubes where the cylindrical part has a mirror symmetry.
Figure 3: An example of a (2, 4) nanotube. The hexagons denoted equally overlap.

The Buckminsterfullerene $C_{60}$ can be viewed as the smallest nanotube of type $(5, 5)$, see Figure 4. It is the smallest nanotube with the caps satisfying the IPR. There are such nanotubes for all types $(i, j)$ with $i + j \geq 11$ and for $(5, 5)$, $(0, 9)$, $(0, 10)$, $(1, 9)$, and $(2, 8)$.

Figure 4: Buckminsterfullerene is the smallest nanotube of type $(5, 5)$.

2 Structural properties of fullerene graphs

As we already mentioned earlier, each fullerene graph has 12 pentagonal faces, all the other faces are hexagons. The distribution of the pentagons determines the shape of the corresponding polyhedron/molecule. If the pentagons are distributed uniformly, the polyhedron is spherical, with the pentagons being the source of the curvature.
2.1 Cyclic edge-cuts in fullerene graphs

Recall that an edge-cut $C$ of $G$ is cyclic if each component of $G - C$ contains a cycle. A graph is cyclically $k$-edge-connected if at least $k$ edges must be removed to disconnect it into two components such that each contains a cycle. For fullerene graphs, Došlić [11] proved the following theorem.

**Theorem 2.1.** Every fullerene graph $F$ is cyclically 5-edge-connected.

Clearly, the result is best possible, since each pentagonal face is separated from the rest of the graph by 5 edges. We say that a cyclic 5-edge cut is trivial, if it separates only one pentagonal face. Kardoš and Škrekovski [38] proved that fullerenes which are not cyclically 6-edge-connected are of unique type.

**Theorem 2.2.** A $(0, 5)$-nanotube is the only fullerene graph with a nontrivial cyclic 5-edge-cut.

A $(0, 5)$-nanotube is depicted on Figure 5. Observe that every nontrivial cyclic 5-edge-cut of a $(0, 5)$-nanotube separates the two caps (containing 6 pentagonal faces each) from each other.

![Figure 5: A $(0, 5)$-nanotube.](image)

Similarly, for each nanotube of type $(i, j)$ there are cyclic $(i+j)$-edge cuts separating the two caps from each other. However, the nanotubes are not the only fullerene graphs with such edge-cuts. An example of a fullerene graph, in which the 12 pentagonal faces are partitioned into four clusters containing 1, 2, 4, and 5 pentagons each, is depicted on Figure 6. It is easy to see that this graph is not a nanotube.
3 Possible stability predictors

Fullerenes were studied from different perspective. The study from graph theoretical point of view has been motivated by a search for invariants that will correlate with their stability as a compound. Banhart et al. [4] observed that molecular structures with sharp edges and facets are less stable than the structures with smooth curvature. In this sense, the more molecule graph is spherical, the more it should be stable.

A number of graph-theoretical invariants were examined as potential stability predictors with various degrees of success [19, 12]. Among the promising candidates for stability predictors are: the bipartite edge frustration, the independence number, the saturation number, the number of perfect matchings, etc. As a result of those investigations, we have achieved a fairly thorough understanding of fullerene graphs and their properties. However, some problems and questions still remain open [8, 43, 25]. Special place among them have several interesting conjectures made by Graffiti, a conjecture making software [18]. Here we consider several of those open questions starting from our results on the fullerene diameters and a recent result on the bipartite edge frustration [17, 20].

3.1 Diameter of fullerene graphs

For large enough fullerene graphs of spherical shape, the diameter $\text{diam}(G)$ (the maximum length of the shortest path between two vertices) is proportional to the radius of the sphere, whereas the number of vertices $n$ is proportional to its surface. Hence, one could expect the diameter of such graphs to be of order $\Theta(\sqrt{n})$. On the
other hand, for a nanotubical fullerene graph of type \((i, j)\), the diameter \(\text{diam}(G)\) is proportional to the length of the cylindrical part of the graph, whereas the number of vertices \(n\) is proportional to the product of the length of the tube and its circumference \(i + j\). In this case, one could expect the diameter of such graphs to be of order \(\Theta(n)\).

A well known result on the degree-diameter problem states that the number of vertices in a planar graph with maximum degree 3 grows at most exponentially with diameter [26].

**Proposition 3.1.** Let \(G\) be a planar graph with maximum degree 3. Then, \(G\) has at most \(2^{\text{diam}(G)+1} - 1\) vertices.

This results gives a logarithmic lower bound on the diameter in terms of the number of vertices.

The logarithmic character of the bound can be attributed to the presence of faces of large size. It would be reasonable to expect that better lower bounds exist for polyhedral graphs with bounded face size.

As mentioned above, fullerene graphs only have pentagonal and hexagonal faces, and this fact can be used to show that the diameter is of order \(\Omega(\sqrt{n})\) indeed.

The following theorem gives the lower bound for the diameter of fullerenes [1].

**Theorem 3.2.** Let \(G\) be a fullerene graph. Then,

\[
\text{diam}(G) \geq \frac{\sqrt{24n - 15} - 3}{6}.
\]

For fullerene graphs with full icosahedral symmetry the diameter has been determined exactly in [3]:

**Theorem 3.3.** Let \(G\) be a \((0, i)\)-icosahedral fullerene graph, with \(i > 0\). Then \(\text{diam}(G) = 6i - 1 = \sqrt{\frac{2}{5}n} - 1\).

**Theorem 3.4.** Let \(G\) be an \((i, i)\)-icosahedral fullerene graph, with \(i > 0\). Then \(\text{diam}(G) = 10i - 1 = \sqrt{\frac{5}{3}n} - 1\).

As we believe that the icosahedral fullerenes have the smallest diameter, it leads us to conjecture the following.
**Conjecture 3.1.** Let $G$ be a fullerene graph on $n$ vertices. Then

$$\text{diam}(G) \geq \left\lfloor \sqrt{\frac{5}{3}n} \right\rfloor - 1.$$  

As far as upper bound is concerned, the nanotubes of small circumference are the extremal graphs, as says the following theorem [1].

**Theorem 3.5.** Let $F$ be a fullerene graph with $n$ vertices. Then,

$$\text{diam}(F) \leq \frac{n}{6} + \frac{5}{2},$$

unless $F$ is a $(0, 5)$-nanotube. In that case, we have $n = 10k$, $k \geq 3$ and

$$\text{diam}(F) = \begin{cases} \frac{n}{3}, & k \in \{3, 4\}; \\ \frac{n}{5} - 1, & k \geq 5. \end{cases}$$

### 3.2 Bipartite edge frustration

The *bipartite edge frustration* of a graph $G$, denoted by $\varphi(G)$, is the smallest cardinality of a set of edges of $G$ that need to be removed from $G$ in order to obtain a bipartite spanning subgraph. Bipartite edge frustration of a fullerene graph $G$ can be efficiently computed by finding a minimum-weight perfect matching in the pentagon-distance graph of $G$ [16]. In the same reference it was shown that $\varphi(G) \geq 6$ for any fullerene graph $G$ and that this bound is sharp. Furthermore, it was shown that the bipartite edge frustration of fullerene graphs with icosahedral symmetry is proportional to the square root of the number of vertices ([16], Proposition 11 and Corollary 12). The numerical computations suggested that it cannot behave worse than that, and prompted the authors to state the following conjecture.

**Conjecture 3.2.** Let $G$ be a fullerene graph with $n$ vertices. Then, $\varphi(G) \leq \sqrt{\frac{12}{5}n}$.  

First, Dvořák, Lidický, and Škrekovski [17] proved a theorem with a weaker multiplicative constant. Later Faria, Klein and Stehlík proved the following theorem, which implies Conjecture 3.2 [20].

**Theorem 3.6.** Let $G$ be a fullerene graph with $n$ vertices. Then, $\varphi(G) \leq \sqrt{\frac{12}{5}n}$. Moreover, the equality holds if and only if $G$ is an $(i, i)$-icosahedral fullerene graph.
The bipartite edge frustration is a very reliable predictor of molecular stability: If \( \varphi(G) \) is close to the upper bound, it means that there is no way to partition the 12 pentagonal faces into six pairs in such a way that the pentagons within the pairs are close to each other.

On the other way, a small value for \( \varphi(G) \) does not say much about the structure of the fullerene graph and about the shape of the molecule. The smallest possible value of \( \varphi(G) \) is attained for any fullerene graph where the 12 pentagonal faces can be partitioned into 6 pairs, adjacent to each other. Those pairs of pentagonal faces can be distributed in a widely varied list of possible configurations, leading to molecules of different shapes, including nanotubes of all possible types.

### 3.3 Independence number

Another invariant tested as a possible stability predictor is the independence number \([19]\). A set \( I \subseteq V(G) \) is independent if no two vertices from \( I \) are adjacent in \( G \). The cardinality of any largest independent set in \( G \) is called the independence number of \( G \) and denoted by \( \alpha(G) \). Independence number of fullerene graphs attracted a lot of attention also in the context of study of independent sets as possible models for addition of bulky segregated groups such as free radicals or halogen atoms [5].

Sharp upper bounds on the independence number of \( n/2 - 2 \) for general fullerenes and \( n/2 - 4 \) for those with isolated pentagons follow by simple counting argument [22]. Lower bounds were gradually improved from (almost) trivial \( \alpha(G) \geq n/3 \) valid for all 3-chromatic graphs to \( \alpha(G) \geq \frac{3}{8}n \) [35] valid for all triangle-free planar cubic graphs. A better lower bound of type \( \alpha(G) \geq \frac{n}{2} - C\sqrt{n} \), for some constant \( C \), was first established for icosahedral fullerenes [32].

This observation was formalized in a pair of conjectures in a recent Ph.D. thesis by S. Daugherty ([8, pp. 96]). The first one states that the minimum possible independence number is achieved on the icosahedral fullerenes that also figure prominently in Conjecture 3.2. The second one [8, Conjecture 5.5.2] states the precise form of the conjectured lower bound. Notice that the constant \( 3/\sqrt{15} \) is exactly one half of the constant \( \sqrt{12/5} \) of Conjecture 3.2.
Conjecture 3.3. Let $G$ be a fullerene graph with $n$ vertices. Then

$$\alpha(G) \geq \frac{n}{2} - 3\sqrt{n/15}.$$

The first result that approaches the conjecture is due to Andova et al. [1].

Theorem 3.7. Let $G$ be a fullerene graph with $n$ vertices. Then

$$\alpha(G) \geq \frac{n}{2} - 78.58\sqrt{n}.$$ 

Finally, using Theorem 3.6, Conjecture 3.3 was confirmed in the following form.

Theorem 3.8. Let $G$ be a fullerene graph with $n$ vertices. Then,

$$\alpha(G) \geq \frac{n}{2} - \sqrt{\frac{3}{5}n}.$$ 

Moreover, the equality holds if and only if $G$ is an $(i,i)$-icosahedral fullerene graph.

Similarly to the bipartite edge frustration, a small value of independence number means that the corresponding polyhedron/molecule is close to being of a spherical shape. On the other hand, a value of $\alpha(G)$ close to $n/2$ does not say much about the structure or the shape of the molecule.

The relations between diameter and the independence number of fullerenes appear in Conjecture 912 of Graffiti [25]. This conjecture was established in [1] for large fullerenes using Theorems 3.5 and 3.7.

Conjecture 3.4. If $G$ is a fullerene graph, then

$$\alpha(G) \geq 2(\text{diam}(G) - 1).$$

Finally, Theorem 3.5 and Theorem 3.8 together confirm the conjecture in the affirmative.

3.4 Upper bound on the smallest eigenvalue

Another invariant investigated for its stability-predicting potential is the smallest eigenvalue of a fullerene graph. In [24] it was proven that the dodecahedron has maximum smallest eigenvalue among all fullerene graphs and it is equal to $-\sqrt{5}$. It
was also shown that the buckminsterfullerene $C_{60}$ has the maximum smallest eigenvalue among all IPR fullerene graphs. This observation lead the authors to state the following conjecture on the smallest eigenvalue of fullerene graphs with at least 60 vertices.

**Conjecture 3.5.** Among all fullerene graphs with at least 60 vertices, the buckminster fullerene has the maximum smallest eigenvalue.

The Conjectures 3.2 and 3.5 are connected via a result on Laplacian eigenvalues from the monograph by Godsil and Royle ([28, pp. 293]).

**Theorem 3.9.** Let $G$ be a graph with $n$ vertices. Then $\text{bip}(G) \leq \frac{2}{7} \mu_\infty(G)$.

Here $\text{bip}(G)$ denotes the maximum number of edges in a bipartite spanning subgraph of $G$ (hence the number of edges in $G$ minus the bipartite edge frustration), and $\mu_\infty(G)$ is the largest Laplacian eigenvalue of $G$.

Recall that the smallest eigenvalue $\lambda_n(G)$ of a 3-regular graph $G$ and the largest Laplacian eigenvalue $\mu_\infty(G)$ of $G$ are related via the following relation ([28, pp. 280]):

$$\lambda_n(G) = 3 - \mu_\infty(G).$$

By plugging this into Theorem 3.9 and noting that $\text{bip}(G) = \frac{3}{2}n - \varphi(G)$ we obtain an upper bound on $\lambda_n(G)$ in terms of the bipartite edge frustration of $G$ of the form $\lambda_n(G) \leq -3 + \frac{4}{n} \varphi(G)$. By taking into account an upper bound on $\varphi(G)$ we immediately obtain the following upper bound on the smallest eigenvalue of a fullerene graph with $n$ vertices [20].

**Theorem 3.10.** Let $G$ be a fullerene graph with $n$ vertices. Then,

$$\lambda_n(G) \leq -3 + 8\sqrt{\frac{3}{5}n}.$$  

### 3.5 Number of perfect matchings

Since all carbon atoms are 4-valent, for every atom precisely one of the three bonds should be doubled. Such a set of double bonds is called a Kekulé structure in a fullerene. It corresponds to the notion of perfect matchings in fullerene graphs: a
matching in a graph $G$ is a set of edges of $G$ such that no two edges in $M$ share an end-vertex. A matching $M$ is perfect if any vertex of $G$ is incident with an edge of $M$.

Let $M$ be a perfect matching in a fullerene graph $G$. A hexagonal face is resonant if it is incident with three edges in $M$. The maximum size of a set of resonant hexagons in $G$ is called the Clar number of $G$.

The computation of the number of perfect matchings in typical fullerene graphs with a small number of vertices [13] indicates that this number should grow exponentially with $n$.

The first general lower bounds for the number of perfect matchings in fullerene graphs were linear in the number of vertices [9, 10, 45]. The best of them asserts that a fullerene graph with $n$ vertices has at least $\left\lceil \frac{3(n+2)}{4} \right\rceil$ different perfect matchings [45].

The number of perfect matchings was proved to be exponential for several special classes of fullerene graphs: (0,5)-nanotubes [42], icosahedral fullerenes [13] or those who are obtained using specific operations [15], before the following general result [37].

**Theorem 3.11.** Let $G$ be a fullerene graph with $n$ vertices that has no non-trivial cyclic 5-edge-cut. The number of perfect matchings of $G$ is at least $2^{\frac{n-380}{61}}$.

The idea of the proof is to find a perfect matching with $(n - 380)/61$ non-adjacent resonant hexagons. For such a perfect matching, any subset of the set of resonant hexagons can be switched to obtain a different perfect matching.

The bound from Theorem 3.11 is far from being optimal. A little is known about the relationship between the number of perfect matchings of a fullerene graph and the shape of the corresponding molecule.

### 3.6 Saturation number

The last invariant considered here, the saturation number, is also related to matchings. The existence of perfect (and hence maximum) matchings in fullerene graphs has been established long time ago, and there are many papers concerned with their structural and enumerative properties [10, 37, 39]. Another class of matchings, the maximal
matchings, have received much less attention so far, in spite of being potentially useful as mathematical models of dimer absorption. A matching $M$ is maximal if it cannot be extended to a larger matching of $G$. The saturation number of $G$, denoted by $s(G)$, is the cardinality of any smallest maximal matching of $G$. The saturation number of fullerene graph was studied in [10, 14], where the following bounds were established.

**Theorem 3.12.** There exists an absolute constant $C$ such that

$$\frac{3n}{10} \leq s(G) \leq \frac{n}{2} - C \log_2 n,$$

for any fullerene graph $G$ with $n$ vertices.

Using the lower bound on the diameter we are able to improve the logarithmic additive correction of Theorem 3.12 and to prove the following result [1].

**Theorem 3.13.** Let $G$ be a fullerene graph with $n$ vertices. Then,

$$s(G) \leq \frac{n}{2} - \frac{1}{4}(\text{diam}(G) - 2).$$

In particular,

$$s(G) \leq \frac{n}{2} - \frac{\sqrt{24n - 15} - 15}{24}.$$  

Still, this is not the best possible bound of the saturation number of the fullerene graphs. In [2] we prove that the saturation number for fullerenes on $n$ vertices is essentially $n/3$.

**Theorem 3.14.** Let $F$ be a fullerene graph on $n$ vertices. Then,

$$\frac{n}{3} - 2 \leq s(F) \leq \frac{n}{3} + O(\sqrt{n}).$$

In order to prove the lower bound of this theorem we used the discharging method. For the upper bound we first used the Theorem 3.6, and obtained a bipartite graph $F'$. Later, we establish that $F'$ is an induced subgraph of a hexagonal lattice, or an induced subgraph of a hexagonal tube (defined as on Figure 3). Then define a maximal matching on $F'$ such that from each hexagon precisely four vertices are covered by the matching.
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