Hamilton Cycles and Paths in Fullerenes†

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It has been conjectured that every fullerene, that is, every skeleton of a spherical trivalent graph whose set of faces consists of pentagons and hexagons alone, is Hamiltonian. In this article the validity of this conjecture is explored for the class of leapfrog-fullerenes. It is shown that, given an arbitrary fullerene F, the corresponding leapfrog-fullerene Le(F) contains a Hamilton cycle if the number of vertices of F is congruent to 2 modulo 4 and contains a long cycle missing out only two adjacent vertices, and thus also a Hamilton path, if the number of vertices of F is divisible by 4.

1. INTRODUCTORY AND HISTORIC REMARKS

Fullerenes are molecular carbon cages, that is, all-carbon 'sphere'-shaped molecules with trivalent polyhedral skeletons, having 12 pentagonal faces and all other hexagonal faces. They are a very important class of molecules, and thousands of patents already exist for a broad range of pharmaceutical, electronic, and other commercial applications. Also, fullerenes generated an outburst of research articles dealing with this line of research both from a chemical and mathematical viewpoint.[1−10] In mathematical language, fullerenes correspond to trivalent (cubic) and 3-edge-connected planar graphs which have, in view of the well-known Euler formula, 12 pentagons and the remaining faces are hexagons. (A graph is 3-edge-connected if at least three edges are needed to be removed in order to disconnect the graph.) With the upsurge in carbon chemistry following the discovery of fullerenes, graph theory has renewed prominence in chemistry as a means of obtaining systematic qualitative information. It is therefore not surprising that many questions about the chemistry of fullerenes together with the methods used to answer these questions find their natural environment in a graph-theoretic context.

The synthesis of fullerenes prompted a natural question as to whether similar carbon structures exist on other closed surfaces. Apart from the sphere only three other surfaces are possible: the torus, the Klein bottle, and the projective plane.[11] Of these only torus-shaped graphite-like carbon structures, also known as *torusenes*, have received enough research attention[12−16] in view of their direct experimental relevance,[17] but also since the other two types are meaningless from a chemical point of view as they do not admit a realization in the Euclidian 3-space. Only two fullerenes, notably the dodecahedron fullerene C_{20} and the buckminsterfullerene C_{60} (that is, the leapfrog-fullerene of C_{20}), are vertex-transitive admitting a transitive group of automorphisms and thus a high degree of symmetry. The situation with torusenes is completely different since all torusenes are vertex-transitive. Consequently, results proved by Thomassen[18] ensure, among others, existence of a Hamilton cycle in an arbitrary torusene. (A Hamilton cycle in a graph is a cycle passing through every vertex.) On the other hand, despite the fact that a long standing conjecture suggests the same should also be true for fullerenes,[19] not much progress has been made in that respect. This conjecture is related to two well-known open problems about Hamilton cycles. The first one is a conjecture due to Barnette[20] regarding the existence of Hamilton cycles in 3-connected planar bipartite graphs, where it is known, for example, that the conjecture holds true when no faces are bigger than hexagons.[21] (By the Euler formula such a graph has 8 quadrangles and the rest are all hexagons.) The second problem, a variation of the first Barnette conjecture and also due to Barnette, asks if every 3-connected planar graph whose biggest faces are hexagons contains a Hamilton cycle. This conjecture has been verified for graphs on at most 176 vertices.[22] Of course, the above-mentioned conjecture about existence of Hamilton cycles in fullerenes is just a special case of this conjecture.

In this article we address the above question about Hamilton cycles in fullerenes for the class of leapfrog-fullerenes. Given a fullerene F we let $|F|$ denote the number of vertices of F. The leapfrog-fullerene Le(F) is obtained from F by performing the so-called leapfrog (tripling) transformation which consists in the truncation of the dual of F. Hence, Le(F) = Trun(Du(F)), and thus $|\mathrm{Le}(F)| = 3|F|$. The object of this article is to prove the following result which is, to the best of our knowledge, the first general result of its kind in the context of fullerenes.

Theorem 1.1. *Let F be a fullerene. Then the leapfrog-fullerene* Le(F) *has a Hamilton cycle if* $|F|$ *(and thus also* $|\mathrm{Le}(F)|$*) is congruent to* 2 *modulo* 4 *and contains a long cycle missing out only two adjacent vertices (and thus contains a Hamilton path) if* $|F|$ *(and thus also* $|\mathrm{Le}(F)|$*) is divisible by 4.*

The arguments used in the proof of theorem 1.1 rely on a method first employed in ref 23 in order to show the existence of Hamilton paths and cycles in a certain class of cubic Cayley graphs. The method is explained in the next section and illustrated on two fullerenes on 30 and 36 vertices and their respective leapfrog-fullerenes. The proof of theorem 1.1 is given in section 3.

† Dedicated to Professor Nenad Trinajstić on the occasion of his 70th birthday.

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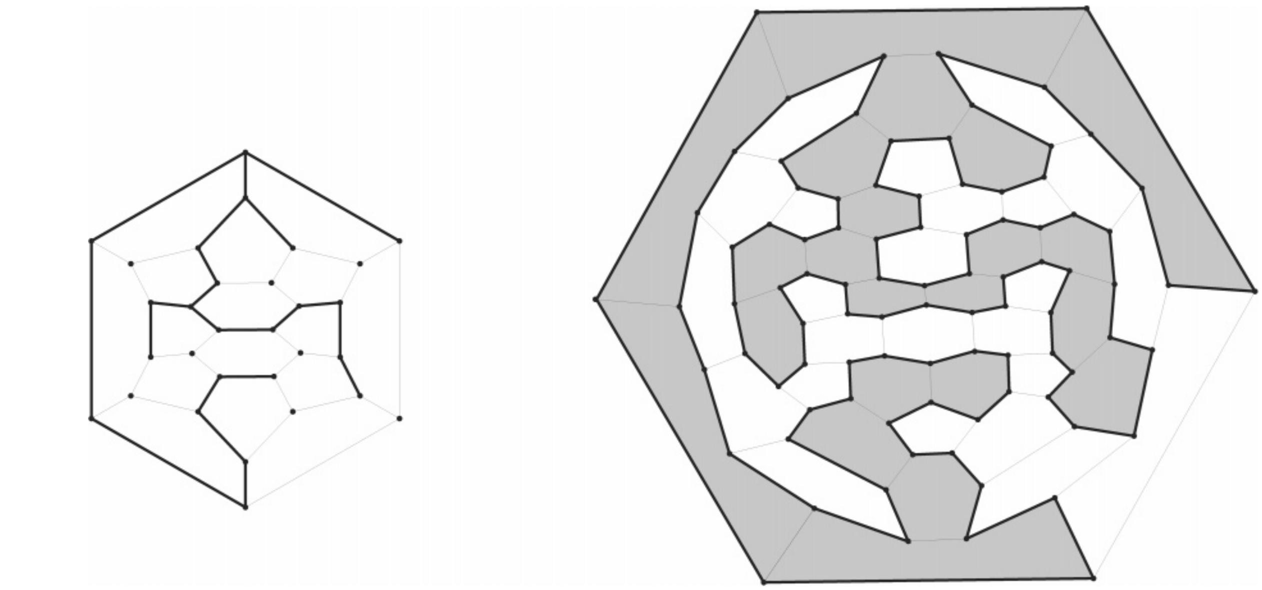

Figure 1. A fullerene on 30 vertices and its leapfrog-fullerene with a Hamilton cycle.

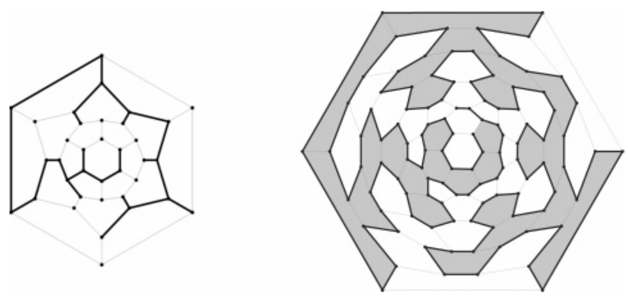

Figure 2. A fullerene on 36 vertices and its leapfrog-fullerene with a Hamilton path.

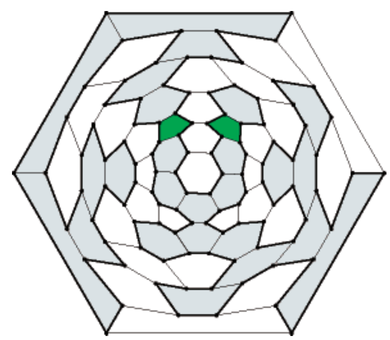

Figure 3. A Hamiton cycle in the leapfrog-fullerene of a fullerene of order 36.

2. EXAMPLES ILLUSTRATING THE METHOD

The method for constructing Hamilton cycles and paths in an arbitrary leapfrog- fullerene consists in identifying, in a given fullerene F, a subset S of vertices V of F inducing a tree, the complement $V \setminus S$ of which has as few edges as possible. In fact, as we shall see, the complement has either no edge when $|F| \equiv 2 (\text{mod } 4)$ or a single edge when $|F| \equiv 0 (\text{mod } 4)$. Hence, in the corresponding leapfrog-fullerene Le(F) this tree gives rise to a tree of hexagonal faces the boundary of which is either a full Hamilton cycle when $|F| \equiv 2 (\text{mod } 4)$ and a long cycle missing only two (adjacent) vertices when $|F| \equiv 0 (\text{mod } 4)$.

Here we give examples of two leapfrog fullerenes, arising from fullerenes on 30 and 36 vertices. Our method gives a Hamilton cycle in the respective leapfrog-fullerene on 90 vertices and a long cycle missing out two adjacent vertices (and therefore a Hamilton path) in the leapfrog-fullerene on 108 vertices. In this example, with a slight modification of the method applied, a Hamilton cycle is produced too.

Example 2.1. In the left picture of Figure 1 we show a fullerene on 30 vertices with an identified induced tree with 22 vertices whose complement is an independent set of 8 vertices. (In view of proposition 3.2 this tree is a maximum such tree.) In the right picture we show the corresponding tree of faces whose boundary gives rise to a Hamilton cycle in the corresponding leapfrog-fullerene on 90 vertices.

Example 2.2. In the left picture of Figure 2 we show a fullerene on 36 vertices with an identified induced tree with 26 vertices whose complement is a set of 10 vertices inducing a single edge. (In view of proposition 3.2, it may be seen that this tree is a maximum such tree.) In the right picture we show the corresponding tree of faces whose boundary gives rise to a long cycle missing only two adjacent vertices in its leapfrog-fullerene. In Figure 3 we show a Hamilton cycle in this leapfrog-fullerene with two of the colored hexagons from the right picture of Figure 2 replaced with two pentagons and one hexagon.

3. PROVING THEOREM 1.1

A graph is *cyclically k-edge-connected* if at least k edges need to be removed in order to disconnect the graph into two components each containing a cycle. The fact that fullerenes are cyclically 5-edge-connected [3,5] will prove to be essential in our construction of Hamilton cycles and paths in leapfrog-fullerenes.

Proposition 3.1. (Došlić [Ref 3, Theorem 2]). *Fullerenes are cyclically 5-edge-connected graphs.*

The concept of cyclic-edge-connectivity is linked to construction of Hamilton cycles and paths through an old result of Payan and Sakarovitch,[25] who considered the so-called cyclically stable vertex subsets (that is, subsets inducing subgraphs with no cycles) in trivalent graphs. The following is an extraction of their main result on a maximum cyclically stable subset of cyclically 4-edge-connected triva-

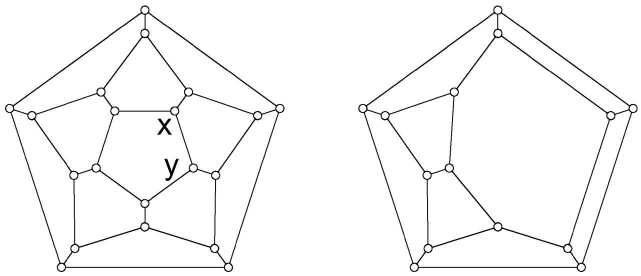

Figure 4. The dodecahedron fullerene on the left and its modified graph on the right.

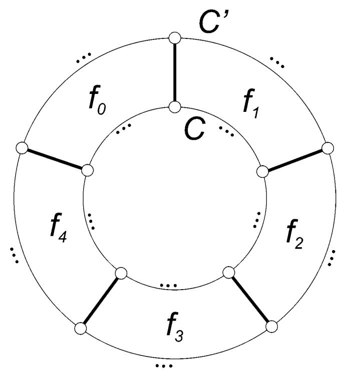

Figure 5. The local structure of a fullerene that admits a ring of five faces.

lent graphs. For a graph X and a vertex subset S we let $X[S]$ denote the subgraph induced by S and all the edges with both endvertices in S. We will use the shorthand notation S^c for the complement $V(X) \setminus S$ of S in the set of vertices $V(X)$ of X.

Proposition 3.2. (Payan, Sakarovitch [Ref 25, Theorem 5]). *Let X be a cyclically* 4*-edge-connected trivalent graph of order n congruent to* 2 *modulo* 4 ($n \equiv 2 \pmod 4$) *, and let S be a maximum cyclically stable subset of* $V(X)$*. Then* $|S| = (3n - 2)/4$ *and* $X[S]$ *is a tree and* S^c *(of cardinality* $(n + 2)/4$*) is an independent set of vertices*.

Before giving a description of cyclically stable subsets in fullerenes of order divisible by 4, an additional concept is needed. Given a connected graph X having at least one vertex of valency different from 2, we let $\mathscr{M}(X)$ denote the multigraph (that is, a graph with multiple edges but no loops) obtained from X by suppressing all vertices of valency 2. Further, if $|V(X)| \geq 6$ and $x,y \in V(X)$ are adjacent, we let $\mathrm{Mod}_{x,y}(X)$ denote the graph $\mathscr{M}(X[\{x,y\}^c])$, the so-called *modified graph* of X relative to the edge xy. In Figure 4 the modified graph of the dodecahedron fullerene C_{20} is shown.

The following result extracted from ref 26 will be used in the proof of our next lemma regarding cyclically stable subsets in fullerenes. For the sake of completeness we outline the proof.

Proposition 3.3. [Ref 26, Lemma 2.3]. *Let F be a fullerene containing a ring R of five faces, and let C and C′ be the inner cycle and the outer cycle of R, respectively. Then either C or C′ is a face, or the five faces of R are all hexagonal*.

PROOF. Let f_0, f_1, f_2, f_3, and f_4 be the faces in the ring R such that f_i is adjacent to f_{i+1}, $i \in \mathbb{Z}_5$. Let T be the set of edges between these five faces (depicted in bold in Figure 5). Clearly, T is a cyclic-5-cutset of F. Moreover, if T is a trivial cyclic-5-cutset then either C or C' is a face. We may therefore assume that T is a nontrivial cyclic-5-cutset and that neither C nor C' is a face.

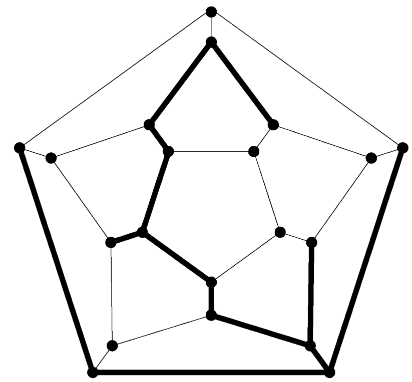

Figure 6. A cyclically stable subset of cardinality 14 in the dodecahedron.

Let S and S', respectively, be the inside of C and the outside of C'. Let $l = l(C)$ and $l' = l(C')$ be the corresponding lengths of cycles C and C'. With no loss of generality, let $l \leq l'$. Depending on whether the five faces are either all pentagonal at the one extreme or all hexagonal at the other extreme or possibly some pentagonal and some hexagonal, we have that

$$15 \leq l + l' \leq 20 \tag{1}$$

Since T is a nontrivial cyclic-5-cutset and C is not a face, we must have that $l \geq 6$.

If $l \in \{6,7\}$, then there either exists one edge or there are two edges having one endvertex in C and the other in S whose deletion disconnects F, contradicting 3-connectedness of F. Therefore $l \geq 8$. However, if $l \in \{8,9\}$ it may be seen, using the fact that the induced graph $F[S]$ is a forest, that the girth of F is smaller than 5, a contradiction. For details see ref 26. Now (1) implies that $l = l' = 10$, and therefore all of the faces f_i, $i \in \mathbb{Z}_5$, on R are hexagons.

Lemma 3.4. *Let F be a fullerene of order n divisible by* 4*. Then there exists a cyclically stable subset S of F of cardinality* $(3n - 4)/4$ *such that* $F[S]$ *is a tree and its complement* S^c *induces a graph with one edge and* $(n - 4)/4$ *isolated vertices*.

PROOF. We choose an arbitrary edge xy in F lying on a pentagon and consider the corresponding modified graph $\mathrm{Mod}_{x,y}(F)$ on $n - 6$ vertices. Since the dodecahedron satisfies the conclusion of this lemma (see Figure 6), we may assume that xy is an edge adjoining a pentagonal and a hexagonal face. There are three possibilities that may occur depending on the local pentagons/hexagons structure of the four faces around the edge xy as shown in Figure 7. In $\mathrm{Mod}_{x,y}(F)$ we again have three possibilities obtained after removal of the six vertices of the set $N(x,y)$ (where $N(x,y) = N(x) \cup N(y)$ denotes the union of neighborhoods of x and y) and after four new edges, shown in bold, have been added in (see Figure 8).

We show first that $\mathrm{Mod}_{x,y}(F)$ is cyclically 4-edge-connected. To see this we just need to analyze each of the above-mentioned three possibilities. We will however only deal with the case of three hexagonal faces and one pentagonal face surrounding the edge xy, depicted in the left picture in Figure 8. The arguments in the remaining two cases are analogous and are thus omitted. Now, suppose on the contrary that $\mathrm{Mod}_{x,y}(F)$ is not cyclically 4-edge-connected and let T be a corresponding cyclic k-edge-cutset, $k \leq 3$, that is, a set the

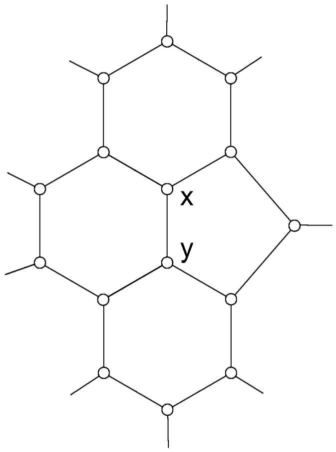

Figure 7. All possible local structures of a fullerene around an edge xy lying on a pentagon and a hexagon.

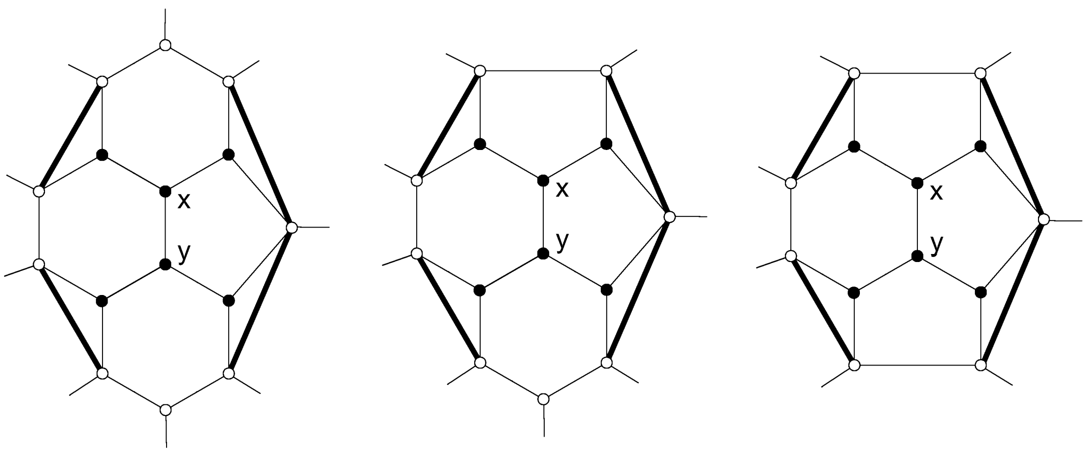

Figure 8. All possible local structures in the modified graph of a fullerene.

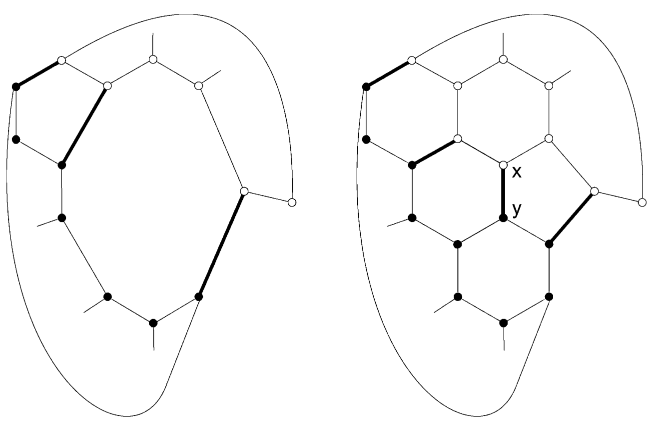

Figure 9. A cyclic 4-edge-cutset.

removal of which decomposes $\mathrm{Mod}_{x,y}(F)$ into two subgraphs each containing a cycle. Let C denote the 9-cycle in $\mathrm{Mod}_{x,y}(F)$ consisting of the four new edges and five old edges. Clearly, each cycle of $\mathrm{Mod}_{x,y}(F)$ contains an even number of edges from T and so, in particular, C has either no edge or two edges from T. If no edge from T is on C, then T is also a cyclic k-edge-cutset of F, $k \leq 3$, contradicting proposition 3.1. We may therefore assume that two edges from T, say e_1 and e_2, are on C. Now e_1 and e_2 are either both old edges (that is also edges of F), both new edges (not edges of F), or one of them is old and the other new.

Suppose first that both e_1 and e_2 are old edges. If they lie on neighboring faces in F, then T together with an appropriate edge (of F) inside C is a cyclic $(k + 1)$-edge-cutset of F, contradicting proposition 3.1. We can therefore assume that e_1 and e_2 lie on non-neighboring faces in F. It follows that we must have an extra edge $e_3 \in T$ (namely if this is not the case, then e_1 and e_2 together with appropriate two edges of F inside C is a cyclic 4-edge-cutset of F). But e_1, e_2, and e_3 together with appropriate two edges of F inside C form a ring of five faces of which at least one is a pentagon, contradicting proposition 3.3.

Similarly, a cyclic 4-edge-cutset may also be found in all other cases. (An example where both e_1 and e_2 are new edges is shown in Figure 9.) These contradictions show that $\mathrm{Mod}_{x,y}(F)$ is indeed cyclically 4-edge-connected.

Now knowing that $\mathrm{Mod}_{x,y}(F)$ is a cyclically 4-edge-connected trivalent graph on $n - 6 \equiv 2(\mathit{mod}\ 4)$ vertices, we may apply proposition 3.2 and deduce the existence of a maximum cyclically stable subset Q of cardinality $(3n - 20)/4$ in $\mathrm{Mod}_{x,y}(F)$ whose complement Q^c is an independent set of cardinality $(n - 4)/4$. Now letting $S = Q \cup (N(x,y)\backslash\{x,y\})$ (and thus $S^c = Q^c \cup \{x,y\}$) we obtain the desired cyclically stable subset of F. This completes the proof of lemma 3.4.

We are now ready to prove theorem 1.1.

Proof of Theorem 1.1. Suppose first that the number of vertices n of F is congruent to 2 modulo 4. By proposition 3.2 there exists a cyclically stable subset S in F of cardinality $(3n - 2)/4$ which induces a tree $F[S]$ and whose complement S^c is an independent set of cardinality $(n + 2)/4$. This tree gives rise to a tree of faces in the leapfrog-fullerene Le(F) whose boundary is a Hamilton cycle in Le(F). This may be seen, for example, by counting the number of vertices on the boundary of this tree of faces. First, the boundary is clearly a cycle. And second, since all faces are hexagonal, the length of this cycle is $6|S| - 2(|S| - 1) = 4|S| + 2 = 3n$, and hence the cycle is indeed a Hamilton cycle.

Now suppose that the number of vertices of F is divisible by 4. By lemma 3.4, there exists a cyclically stable subset S in F of cardinality $(3n - 4)/4$ which induces a tree $F[S]$ and whose complement S^c induces a graph with one edge and $(n - 4)/4$ isolated vertices. Again, a simple counting argument gives us that the boundary of the corresponding tree of faces in the leapfrog-fullerene Le(F) is a cycle of length $3n - 2$ missing out two adjacent vertices in Le(F). Hence the leapfrog-fullerene Le(F) also has a Hamilton path. Note that these two adjacent vertices in Le(F) are endvertices of a common edge of two adjacent hexagons in Le(F) corresponding to the two vertices in S^c inducing the only edge in $F[S^c]$.

4. CONCLUSIONS AND FUTURE DIRECTIONS

In this paper we have used certain graph-theoretic information available on fullerenes, such as their cyclic 5-edge-connectivity and the fact that fullerenes have large induced trees. This has allowed us to construct Hamilton cycles when the order n of a leapfrog-fullerene is congruent to 2 modulo 4 and cycles of length $n - 2$ (and hence also Hamilton paths) when n is divisible by 4. It is quite possible that with some modification (for example, by using two pentagonal faces in the corresponding tree of faces) these methods could give us a full Hamilton cycle in leapfrog-fullerenes in the latter case too. The approach used in this paper could be further explored, say in the context of other classical fullerenes transformations, such as chamfering (quadrupling)[8] and capra (septupling) transformations.[8] In particular, is it reasonable to expect that these methods could be used to show existence of Hamilton (or large) cycles in camfering-fullerenes and capra-fullerenes?

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