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Chemical Physics Letters 428 (2006) 386–393
CHEMICAL PHYSICS LETTERS
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Growing fullerenes from seed: Growth transformations of fullerene polyhedra

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Received 31 May 2006; in final form 11 July 2006
Available online 20 July 2006

Abstract

A cut-and-paste approach using a family of structurally similar 'growth patches' (pairs of non-isomorphic patches with the same boundary but containing different numbers of vertices) allows formal construction from a C_{24} seed of all fullerene isomers with up to at least 200 atoms. Algorithmic and chemical implications are discussed.

1. Introduction

A perennial problem in the chemistry and physics of fullerenes is the question of how these organised cage structures emerge from chaotic low-nuclearity carbon vapour [1,2]. Various particular mechanisms for carbon ingestion/extrusion [3] and isomerisation/annealing [4] have been proposed and assessed by comparison with experimental data and quantum mechanical calculations [5,6]. A parallel line of investigation involves the use of graph theoretical techniques to catalogue the mathematically possible fullerene structures [7,8] and their interconversions, based on assumed sets of rules for construction and transformation [9–14]. The present study extends this approach, exploring the ways that fullerenes can be formally generated from 'seed' polyhedra, using either a predefined set of graph transformations or a set that is restricted by a cost function intended to mimic the energetics of bond rearrangement and carbon insertion. The classical definition of a fullerene as a carbon cage whose skeleton is a trivalent polyhedron with hexagonal and (12) pentagonal faces will be used. It will be shown, for example, that a family of transformations based on the Endo–Kroto C_2 insertion mechanism [3] gives access to *all* isomers of *all* fullerenes up to C_{200} from a C_{24} seed.

2. The patch-replacement approach

These results are obtained using the notion of patch-replacement, which can represent transformations between fullerenes of the same carbon count (isomerisation) or of different carbon counts (growth). Systematic catalogues of isomerisation and growth transformations, ordered by patch boundary and pentagon count, are available [15,16]. The qualitative idea behind patch-replacement is that we can imagine going from one fullerene graph to another by a 'cut-and-paste' operation: take a patch defined by a boundary cycle of edges and vertices and replace it by another interior compatible with the same boundary. Two non-isomorphic patches with the same boundary and number of vertices, or two copies of a patch with a boundary that has a larger symmetry group than the patch itself, are called an *isomerisation pair*. Two patches with the same boundary and different numbers of vertices

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[1] Supported by Royal Society/Wolfson Research Merit Award.

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doi:10.1016/j.cplett.2006.07.040

are called a *growth pair* [15,16]. We distinguish two possible operations: on the one hand, operations where the boundary of the patch may not contact itself in the fullerene and, on the other, operations where the boundary circuit may contact itself, but cannot intersect itself (Fig. 1). Fig. 2 shows the simplest isomerisation and growth pairs: the Stone–Wales patch [4], with which pentagons may be moved around within a suitable fullerene graph, and the Endo–Kroto patch [3], with which a suitable fullerene graph may grow. Both have boundaries that are simple circuits of 12 vertices and 12 edges.

We will say of two fullerenes related by a growth transformation that the larger is *reducible* to the smaller, implying that one can be formally obtained from the other by the cut-and-paste process. A mathematical advantage of the patch/boundary picture is that it might lead to an efficient fullerene generator. If all fullerenes *could* be grown from the smallest graph that contains hexagons – the unique two-hexagon C_{24} cage – using a small repertoire of patches, the construction part of the problem – that of complete generation of objects, staying within the class – would have been solved, and it would remain only to deal with isomorphism rejection, which could be done by standard techniques [18,19].

Against this optimistic view, it is known that an infinite *number* of growth operations would be needed to obtain all fullerenes. This is a consequence of the fact that in a patch with at most one pentagon, the boundary determines the number of faces [17]. So a growth patch contains at least two pentagons and as there is no upper bound on the size of a smallest patch in a fullerene containing two pentagons (we can think of larger and larger icosahedral fullerenes), it is clear that an infinite number of different patches will be needed to reduce these fullerenes.

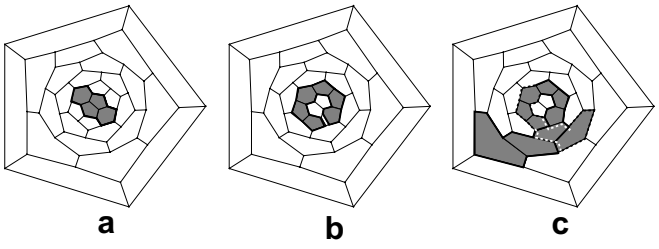

Fig. 1. Allowed patch boundaries are: (a) simple circuits, (b) in some cases circuits with repeated edges/vertices, but never (c) circuits with self-intersections.

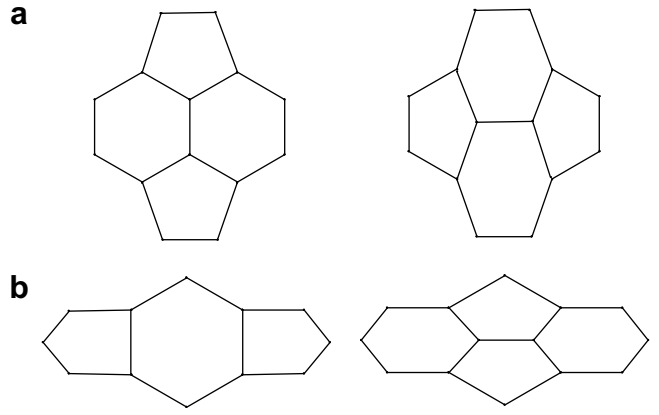

Fig. 2. The two simplest mechanisms for interconversion of fullerene polyhedra: (a) the Stone–Wales isomerisation patch [4] and (b) the Endo–Kroto C_2 insertion patch [3].

It is an open question whether a combination of a finite number of isomerisation and growth steps can access all fullerenes. This question could be answered in the negative if it could be shown that isomerisation patches with less than two pentagons cannot occur as parts of fullerenes. So far, no isomerisation patch is known that has fewer than two pentagons and can occur in a fullerene.

We associate to every patch a cyclic sequence of zeros and ones in the following way: a boundary vertex of a patch that has an edge going inward (or equivalently: has degree 3 in the patch) is assigned a 1 and a boundary vertex that has an edge going outward into the rest of the fullerene (or equivalently: has degree 2 in the patch) is assigned a 0. Following the boundary of the patch we obtain a cyclic sequence of 0s and 1s. We consider two boundaries to be similar if one can be obtained from the other by repeating a certain fixed part of the sequence. To be exact: if for given $l \in \mathbb{N}$ and a_i, b_i words in an alphabet of symbols 0 and 1, both can be written in the form $\prod_{i=1}^{l} a_i b_i^k$ for some $k \in \mathbb{N}$. (k may be different for both patches.)

We define a set of patches to constitute a family, if: (a) there is a formula $\prod_{i=1}^{l} a_i b_i^k$ such that every boundary can be written in that way and (b) for every k greater or equal to a given k_0 there is at least one pair of patches with boundary $\prod_{i=1}^{l} a_i b_i^k$ that forms a growth pair.

Another open question is then whether a single infinite 'family' of growth transformations will suffice to reach all fullerenes from a finite number of 'seed' fullerene graphs.

3. Chemical aspects

From the chemical viewpoint, there are some more limited practical questions. Can all fullerenes up to some reasonable number of vertices be reached with a finite set of patches or family of patch types? Not all patches are equally attractive as candidates for physical processes of interconversion as they involve widely differing amounts of activation energy. Supposing that the energy cost of a growth transformation can be defined, at least approximately, by a graph theoretical property of the patch pair, it is an interesting question to investigate accessibility of fullerenes subject to a given cost function. What fullerenes are accessible if a given total energy is available per growth step? Physically reasonable cost functions are likely to encourage use of small steps over steps that insert large fragments.

One consideration that affects the use of the published catalogue of growth patches [16] for exploration of reducibility in the present sense is that the catalogues of growth and isomerisation patches were compiled with a notion of

minimality in mind. The minimality criteria applied in generation of the catalogue eliminated patches that were 'essentially' the same as one already included apart from some 'spectator' face. For example, the Stone–Wales/pyracylene transformation [4] is 'essentially' the same transformation whether or not extra faces are added on the boundary of the patch. Removal of such 'spectator' faces can either increase or decrease the boundary length and so it is necessary to check explicitly that a given search for reducibility in the sense of the present Letter takes into account all relevant patches.

Cost functions might also differentiate between rearrangement of pentagon–pentagon, pentagon–hexagon and hexagon–hexagon edges, recognising that such edges have different probabilities of bearing formal single and variable bonds in the most appropriate Kekulé structures for fullerene graphs, and that placing of double bonds in the pentagons and hexagons may carry different steric and electronic penalties and benefits.

4. Methods

The simplest (shortest-boundary) growth patch is the Endo–Kroto patch [3], which has a maximum local C_{2v} symmetry, consists of one hexagon sandwiched between two pentagons, and on replacement within its 12-vertex boundary, leads to incorporation of two extra vertices in the fullerene graph. Two natural extensions (see Fig. 3) are: (i) the untwisted (C_{2v}) patch with $k+1$ hexagons sandwiched between the two pentagons and (ii) the twisted (C_2) patch with two pentagons attached to a straight strip of $k+1$ hexagons in one of two possible enantiomeric versions [7,16]. Both series have the Endo–Kroto patch as the smallest element in the set.

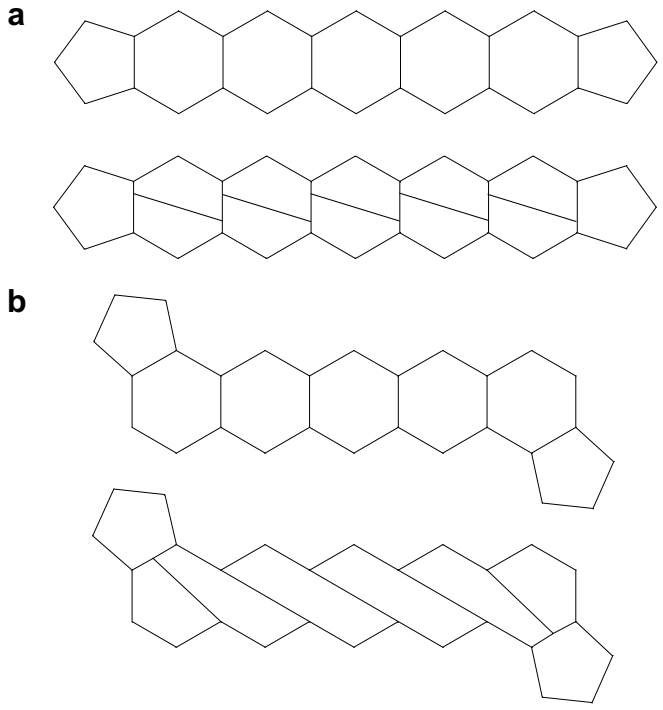

Fig. 3. Extensions of the Endo–Kroto patch. Patch boundaries with: (a) C_{2v} symmetry and (b) C_2 symmetry. In case (a) each extension inserts two new vertices in every hexagonal face of the patch while in case (b) each extension inserts two new vertices in total.

A computer program was written to find patch pairs and series, e.g., of types (a) ('C_{2v} Endo–Kroto') and (b) ('C_2 Endo–Kroto') in fullerene isomers C_n, and to determine reducibility of the isomer subject to possible further restrictions.

Input to the described computer program is a set of fullerenes, generated by *fullgen* [8], either a description of the boundary and the minimal number of vertices that a patch with that boundary can have, or a set of patches so that the program can determine these values itself. For every input fullerene the computer program first checks for every directed edge in the fullerene whether the given boundary, defined as right and left turns, closes without intersections and – in case that they are forbidden – also without self contacts when starting from this edge. In case such a boundary is found, the number of vertices in the interior is counted and in case it is larger than that of the smallest patch with the given boundary, the fullerene is declared as reducible. Since in some cases this allows for a large number of bigger patches, there is also the option to give a maximal number of vertices of the patch. This way it is often possible to restrict the search enough that only one patch with the given boundary – the second smallest that is possible – can give rise to reducibility of the fullerene. Furthermore there is also an option that counts the number of edges in the interior of the patch found in the fullerene and in the smallest possible patch with the same boundary and accepts only cases where the sum of these is not too large. In fact we never have to look for isomorphism of the patches found in the fullerene and the patches given.

In the case of fullerenes obeying isolated-pentagon rule (i.e., the IPR-fullerenes) there is also an option that makes the program check that at certain places outside of the boundary of the patch found in the fullerene there are no pentagons. This can guarantee that the fullerene is reducible to another, smaller IPR-fullerene. This option is not compatible with a boundary that contacts itself, so all results given for IPR-fullerenes are for the case of forbidden boundary contact.

In order to reduce the probability of programming errors, we implemented the approach independently once in C and once in Java and computed the vast majority of the results given here independently by both programs.

Using the results of the programs, it was possible to construct connection tables for the isomers under various assumptions for the set of allowed transformations, in other words by playing out various 'growth games'.

5. Results

C_{20} is an irreducible fullerene under all possible patch transformations, since it is the smallest possible fullerene.

C_{24}, as the first non-C_{20} fullerene isomer, is irreducible under all variations of the Endo–Kroto patch transformation, but can be reduced using a replacement patch with 4 pentagons.

The C_2 Endo–Kroto patch series is found to be remarkably efficient in generating larger fullerenes from smaller – even when the boundary is prohibited to contact itself and only the pair consisting of the two smallest patches for every boundary is considered. Starting from a C_{24} 'seed', *all* fullerene isomers of up to $n = 200$ are generated, with three exceptions: the D_{3d} isomer of C_{44} known in the fullerene spiral notation [7] as 44:3 (Fig. 4) and one isomer of each of C_{164} and C_{200}. It is easy to see that 44:3 cannot be generated by a type (b) patch because, due to the fact that the pentagons involved must have 2 hexagonal neighbours and due to the high symmetry, only two possibilities have to be checked. Thus, all fullerenes from C_{26} to C_{200}, with just three exceptions, can be generated from the one 'seed' of C_{24}. If one allows the boundary to contact itself, *all* fullerenes from C_{26} to C_{200} – without exception – can be generated from the one 'seed' of C_{24}.

The symmetric C_{2v} Endo–Kroto patch can also be used to grow fullerenes from C_{24}, and, as Table 1 shows, it does fairly well, but is not as efficient as the twisted version. The irreducible fullerenes (even when contacts of the boundary are allowed) include an infinite family of exceptions in the cylinders with hemi-dodecahedral caps (the isomers n:1 with $n = 10k$ vertices) (30:1 (D_{5h}), 40:1 (D_{5d}), 50:1 (D_{5h}) ...). Furthermore there are various sporadic examples of irreducible fullerenes already for small vertex numbers: (36:13 (D_{3h})), (38:9 (D_3)), (40:40 (T_d)), (52:94 (D_{2d})), (56:459 (D_{2h})), (56:622 (T_d)), (56:910 (D_3)), (60:208 (C_{2v})) until at 200 vertices there are 286 irreducible isomers.

There is an appealing simplicity in restricting the allowed patches to a single structural family, but this is certain to result in the need for larger and larger patches as n grows. Physically, growth processes will be limited by the available activation energy, as well as the need for atoms involved in the insertion to be present in the right place at one time. On these grounds, it seems sensible to run the same growth game, but with imposition of a cost function. If the reaction is concerted, and boundary relaxation effects are not dominant, the more bonds are made and broken, the higher the activation energy we can expect. This assumption can be formalised by writing the cost as proportional to the sum of numbers of old edges broken and new edges formed, where the proportionality factor is some typical bond energy. The Endo–Kroto patch then has the smallest cost, with $2 + 5 = 7$ units, the C_{2v}-symmetry pair with h-hexagons in the smaller patch has a cost of $5h + 2$ units and the C_2-symmetry pair with h-hexagons in the smaller patch has a cost of $2h + 5$ units.

This cost function requires a definition of the minimum size of a patch. The inclusion of a spectator face would increase the apparent cost as it would increase the number of interior edges both before and after transformation. A well defined minimum replacement cost as a function of the boundary length can be worked out. In the optimal case, the minimal patch is empty of interior vertices, and

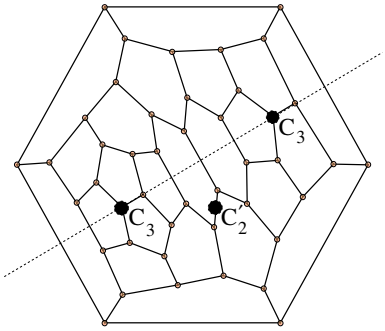

Fig. 4. The irreducible D_{3d} isomer of C_{44}.

Table 1
Numbers of fullerenes irreducible by the C_{2v}-symmetry Endo–Kroto family

Number of atoms	Irreducible (A)	Irreducible (B)
20	1	1
24	1	1
26	0	0
28	0	0
30	1	1
32	0	0
34	0	0
36	1	1
38	1	1
40	3	2
42	0	0
44	0	0
46	0	0
48	1	0
50	1	1
52	1	1
54	1	1
56	4	3
58	0	0
60	2	2
62	2	2
64	1	0
66	0	0
68	3	2
70	2	2
72	3	2
74	0	0
76	1	1
78	1	1
80	5	4
82	0	0
84	3	3
86	2	2
88	5	3
90	1	1
92	4	4
94	2	2
96	3	2
98	1	1
100	7	7

In (A) patches with touching boundary are forbidden, in (B) they are allowed.

Table 2
Numbers of irreducible fullerenes for some reductions with low cost

Number of atoms	Number of fullerenes	Cost 7	Cost 9	Cost 11	Cost 12	Cost 13	Cost 14
20	1	1	1	1	1	1	1
22	0	0	0	0	0	0	0
24	1	1	1	1	0	0	0
26	1	0	0	0	0	0	0
28	2	0	0	0	0	0	0
30	3	1	0	0	0	0	0
32	6	2	1	0	0	0	0
34	6	0	0	0	0	0	0
36	15	2	0	0	0	0	0
38	17	2	0	0	0	0	0
40	40	3	1	0	0	0	0
42	45	0	0	0	0	0	0
44	89	6	2	1	1	1	0
46	116	3	0	0	0	0	0
48	199	4	1	0	0	0	0
50	271	4	2	1	1	0	0
52	437	8	1	0	0	0	0
54	580	6	0	0	0	0	0
56	924	20	6	2	1	1	1
58	1205	7	0	0	0	0	0
60	1812	19	4	2	1	1	1
62	2385	24	1	1	1	0	0
64	3465	28	1	1	0	0	0
66	4478	18	0	0	0	0	0
68	6332	54	7	4	1	1	1
70	8149	42	1	1	1	1	1
72	11190	75	6	1	0	0	0
74	14246	72	1	1	1	1	1
76	19151	116	5	1	0	0	0
78	24109	137	2	1	0	0	0
80	31924	208	16	8	2	2	2
82	39718	186	1	0	0	0	0
84	51592	311	14	1	0	0	0
86	63761	370	7	5	1	1	1
88	81738	487	16	1	0	0	0
90	99918	571	5	1	1	1	1
92	126409	805	34	5	1	1	1
94	153493	916	6	0	0	0	0
96	191839	1263	41	3	0	0	0
98	231017	1583	7	1	1	1	1
100	285914	2122	47	7	1	1	1
102	341658	2495	17	0	0	0	0
104	419013	3434	95	11	1	1	1
106	497529	4223	26	0	0	0	0
108	604217	5597	108	5	0	0	0
110	713319	6796	42	8	2	2	2
112	860161	8983	165	2	0	0	0
114	1008444	11087	70	8	0	0	0
116	1207119	14559	283	20	1	1	1
118	1408553	17649	121	2	0	0	0
120	1674171	22682	416	16	1	1	1
122	1942929	28260	161	6	1	1	1
124	2295721	35977	663	11	0	0	0
126	2650866	43176	314	9	0	0	0
128	3114236	55157	1100	49	1	1	1
130	3580637	67677	548	11	1	1	1
132	4182071	83501	1517	35	0	0	0
134	4787715	101495	862	35	1	1	1
136	5566948	125987	2548	31	0	0	0
138	6344698	151693	1322	24	0	0	0
140	7341204	185535	3842	115	3	3	2
142	8339033	223705	2549	36	0	0	0
144	9604410	271856	5609	122	0	0	0
146	10867629	323161	3787	75	1	1	1

(*continued on next page*)

Table 2 (*continued*)

Number of atoms	Number of fullerenes	Cost 7	Cost 9	Cost 11	Cost 12	Cost 13	Cost 14
148	12469092	392534	9075	122	0	0	0
150	14059173	464384	5900	120	2	1	1
152	16066024	555529	13349	373	3	1	1
154	18060973	660162	10120	112	0	0	0
156	20558765	783149	19378	464	0	0	0
158	23037593	920356	14966	365	1	1	1
160	26142839	1092662	30133	529	2	1	1
162	29202540	1274238	22869	386	1	0	0
164	33022572	1500402	43108	1334	3	2	1
166	36798430	1751287	36618	553	0	0	0
168	41478338	2043783	62716	1664	6	2	0
170	46088148	2370495	52983	1282	4	3	2
172	51809018	2766500	92871	2161	4	2	0
174	57417255	3187225	78746	1784	2	0	0
176	64353257	3692948	131149	4318	3	2	1
178	71163435	4254187	118443	2309	3	0	0
180	79538725	4900921	186009	5810	10	5	1
182	87738289	5610804	168390	4942	14	5	1
184	97841157	6456333	266632	7853	19	6	0
186	107679684	7359855	241807	6427	7	2	0
188	119761030	8418313	367636	13947	29	11	1
190	131561725	9584710	348999	9479	15	2	1
192	145976654	10916720	511262	18445	45	10	0
194	159999441	12365760	483018	16710	28	5	1
196	177175662	14064380	708830	25302	57	20	0
198	193814634	15875462	674914	23308	63	13	0
200	214127713	17960189	958543	41825	106	24	2

so the number of edges of the initial patch to be removed is just half the number of entries '1' in the boundary code, whereas the number of edges to be inserted in the final patch is the same *plus* 3/2 times the number of internal vertices of the final patch (of which there are at least two). This implies that for a patch of boundary length b that includes p pentagons, the minimal cost of inserting two vertices is $(b+p)/2$ in the case of a starting patch with no interior vertices (in which case $b-(6-p)$ is divisible by 4) and $(b+p)/2+3$ in the case of a starting patch with a non-empty interior. This gives us an upper bound for the boundary lengths of the patches that have to be examined when searching for growth pairs with a given cost.

Note that we use only patch pairs from the catalogue published in [16], and owing to the notion of *reducibility* of patches, testing for reducibility with cost c does not include testing for a pair with cost $c' < c$ that becomes a pair of cost c by adding spectator faces. So, it is important to test always for cost *at most* c for a certain c. Results of the tests on fullerenes up to 200 vertices are shown in Table 2.

The number of pairs with cost at most k ($k = 7, 8, \ldots 14$) are: 1, 1, 2, 2, 6, 12, 50, 55. In fact, all the fullerenes up to 200 vertices that were found to be reducible by the full list of 55 pairs of cost at most 14 were already reducible by a small subset of this list (with only 8 pairs). We did not check systematically for a subset of minimal size, so it is possible that an even smaller set exists. The set of eight consists of the following pairs (using the numbers in the catalogue [16]): cost 7: G2.12.1.1,2; cost 9: G2.16.2.1,2; cost 11: G2.20.3.1,2; cost 13: G2.24.8.1,2 (these are the four smallest pairs in the C_2 Endo–Kroto family); cost 12: G2.16.3.1,2 (this is the second smallest pair in the C_{2v} Endo–Kroto family); cost 12: G3.15.1.1,2; cost 12: G4.14.1.1,2; cost 14: G2.20.5.1,2.

We can also look at growth of isolated-pentagon fullerenes. If these fullerenes are typically of lower energy than those with adjacent pentagons, it seems reasonable on physical grounds to search for growth patches that will stay within the isolated-pentagon class. The twisted and untwisted Endo–Kroto families are not particularly efficient in this regard, leaving substantial numbers of irreducible isomers, even below 100 vertices. However, this situation changes significantly if one does not require the larger patch in the reduction process to be an element of the family, but allows it to be an arbitrary patch with the same boundary, still replacing it with the smaller of the corresponding Endo–Kroto pair. As an example, for the patches G2.24.8.1-6 in [16], the first two form a pair in the family of twisted Endo–Kroto patches, but there are also four larger patches with the same boundary. *Extended* twisted Endo–Kroto reducibility would consider five possible reduction pairs for this boundary: G2.24.8.1 paired with each of G2.24.8.2-6. Table 3 gives the numbers of irreducible IPR-fullerenes up to 150 vertices for both notions of reducibility. The fullerenes that are neither reducible with extended EK (C_2) nor extended EK (C_{2v}) include all IPR isomers on up to 74 vertices, and many (20 out of 47) of the IPR isomers from 76 to 84 vertices, but then appear to be petering out in the remainder

Table 3
Numbers of IPR-fullerenes that are irreducible within the class of IPR-fullerenes under different notions of reducibility

Number of atoms	Irreducible EK (C_{2v})	Irreducible extended EK (C_{2v})	Irreducible EK (C_2)	Irreducible extended EK_2	Irreducible extended EK (C_{2v}) or EK (C_2)
70	1	1	1	1	1
72	1	1	1	1	1
74	1	1	1	1	1
76	1	1	2	2	1
78	5	5	4	2	2
80	6	6	5	5	4
82	8	8	7	7	7
84	21	20	16	7	6
86	16	16	5	0	0
88	18	17	8	1	0
90	20	18	12	0	0
92	29	27	18	3	0
94	18	17	5	2	1
96	28	26	16	0	0
98	19	15	12	2	0
100	23	18	14	0	0
102	31	26	8	0	0
104	38	23	9	1	0
106	28	14	8	0	0
108	55	18	10	0	0
110	21	10	0	0	0
112	28	7	6	0	0
114	28	9	5	1	1
116	36	10	4	0	0
118	28	8	4	0	0
120	49	15	9	2	1
122	37	12	2	0	0
124	60	15	3	0	0
126	42	8	3	0	0
128	57	11	2	0	0
130	42	2	2	0	0
132	89	5	9	0	0
134	50	0	5	0	0
136	63	1	2	0	0
138	56	2	7	0	0
140	61	6	6	0	0
142	42	5	2	0	0
144	87	10	5	1	0
146	71	6	1	0	0
148	99	4	4	0	0
150	105	6	4	0	0

The *extended* reducibility refers to reduction from any patch of given boundary to the smaller EK patch with the boundary.

of the range to 150 vertices and beyond: the sporadic irreducible higher fullerenes in this category are: a C_{3v} isomer of C_{94} with nine pentagons in the same relation as in C_{80}, a D_{3h} cylinder with a C_{60} cap (C_{114}), D_{5d} cylinders with a C_{80} cap (C_{120}, C_{160}, C_{200}), and a D_{3d} cylinder with a C_{60} cap (C_{168}).

6. Discussion

The calculations presented here have shown that all the fullerenes within present range of chemical characterisation can be generated, formally, from the smallest hexagon-containing fullerene, C_{24}, using a single family of growth patches. Specifically, all isomers of all fullerenes with up to 200 atoms are accessible through a family that generalises the simplest proposal for growth, the Endo–Kroto patch [3]. Moreover, the admittedly crude approach of a graph theoretically defined cost function suggests that a large percentage of the full set of isomers will be accessible with involvement of less than a dozen breaking-and-forming bonds.

Two natural questions arise in relation to the growth game as described so far. We have found a structurally similar family of patches that generates all fullerenes to C_{200}. The practical question is: Is the whole family required to generate these fullerenes, or is there some structurally varied but smaller set of patches that would do the same job for fullerenes up to some limiting number of atoms? The mathematical question is: If the twisted-EK family is retained as the sole set, can we generate *all* fullerenes?

The first question is open. The second is easily answered in the negative, since it is possible to devise a counterexam-

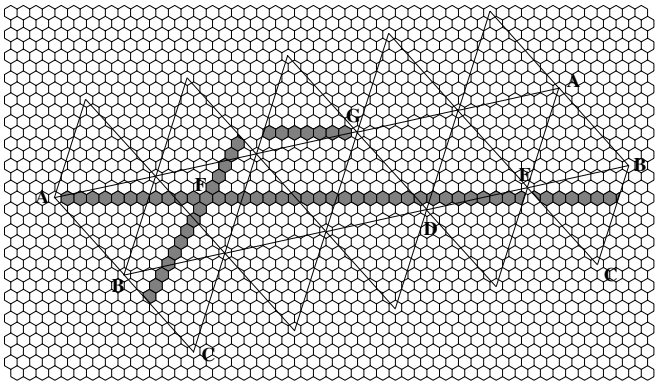

Fig. 5. A fullerene with 1340 atoms that cannot be constructed using only growth operations from within the C_2-symmetry family. Vertices of the net represent pentagon sites. A path though opposite edges of hexagons, starting from the pentagon at A on the left of the diagram, traverses the net, narrowly missing pentagons at points D and E, leaves the net between B and C on the right of the diagram, re-enters at the left, and intersects with itself at point F before eventually hitting a pentagon at point G. By symmetry, all such paths self-intersect. This is the smallest such fullerene with icosahedral symmetry; it is an open question whether there are smaller fullerenes of other symmetries that cannot be so constructed.

ple, a fullerene that cannot have been constructed from any smaller fullerene using any twisted EK patch. The smallest counterexample known to us is the icosahedral fullerene with Coxeter coordinates [20] (7, 2) and 1340 atoms. An unfolding of it is depicted in Fig. 5. The irreducibility can be seen by observing that every path leaving a pentagon and going straight through every hexagon intersects itself before entering the next pentagon. So we will not be able to find a subgraph isomorphic to the larger patch in one of the pairs in the family.

From a pragmatic point of view, it becomes important to know the size of the smallest counterexample. If no such example occurs before, say, 250 or 300 atoms, then the twisted-EK series could give an efficient way to generate all fullerenes in the range where consideration of the full set of isomers is still a practical proposition.

Work on refining the counterexample and on selection of an optimal set of patches is in progress.

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