

Analytical Expressions for the Moments and Characteristic Polynomials of Fullerenes Containing Isolated Pentagons

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Received: April 29, 1993*

On the basis of the molecular fragment method, the analytical formulas for the moments (μ_k) and the coefficients of characteristic polynomials (a_k) were derived up to $k = 14$ for fullerenes containing isolated pentagons (FCIPs). It is interesting that the first 11 moments and the coefficients in the characteristic polynomials of isomers of these fullerenes are identical as they depend only on the number of carbon atoms. For $k \geq 12$ additional variables have to be introduced to express the moments and the coefficients in the characteristic polynomials.

Introduction

Stimulated by the exciting discovery of the unusually stable buckminsterfullerene C_{60} cluster,¹ numerous studies have been made on the fullerene clusters and their properties in recent years.²⁻¹⁹ Theoretical calculations of many physical and spectroscopic properties of fullerenes have been carried out including electronic energies, aromaticities, characteristic polynomials, optical spectra, vibrational modes, and the electric and magnetic properties. The fullerene cages with a metal atom inside have also been investigated as viable candidates for superconducting materials. The buckminsterfullerene cluster is also considered to be of astrophysical importance.

The graphite-arc generated carbon soot was found to contain macroscopic quantities of C_{60} and C_{70} as well as other fullerenes. Several spectroscopic studies have been made on chromatographically separated toluene extract of carbon soot. Up to now in addition to the most abundant C_{60} fullerene, C_{70} , C_{72} , C_{76} , C_{78} , C_{84} , and other fullerenes have been isolated. Some of the higher fullerenes exhibit different isomeric forms and were found to exist as mixtures of several isomers.

Mathematical and topological studies on fullerenes are on the increase in recent years.^{2-4,7-8,17-19} The earlier studies focused on group theoretical and graph theoretical properties of the C_{60} buckminsterfullerene as well as the use of the conjugated circuit methods³⁹ to estimate the relative stability of the C_{60} fullerene and graphite. Subsequently several studies were made on the enumeration of isomers of fullerenes, substituted fullerenes, theoretical prediction of ^{13}C NMR spectra of fullerenes, spectral moments of the C_{60} buckminsterfullerene and so on. The nuclear spin statistics of several fullerenes have also been considered recently.¹⁷⁻¹⁹ The characteristic and matching polynomials of several fullerenes have been obtained, and the first few coefficients have been analyzed.²⁰⁻²²

As dominant graph invariants, the moments and characteristic polynomials of graphs have played an important role in chemical graph theory and its applications.²⁰⁻³⁷ Several useful quantities have emerged from these invariants which facilitate characterization of fullerenes and preliminary for the estimation of relative stabilities and reactivities of such species. The moment method was proven to be an alternative technique to explore the relationship between molecules and the solid state materials.³² Also the moment method facilitated rapid evaluation of energies. Recently, some authors have been engaged in the investigation of moments and the characteristic polynomials of fullerene cages.

According to the generalized Hückel rule,³⁴ the $(4n + 2)$ th moment stabilizes while the $4n$ th moment destabilizes π -electron

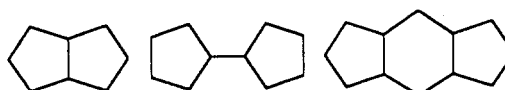


Figure 1. Graph segments which contribute to 8th and 12th moments.

molecules, where $n = 0, 1, 2, \dots$. This is suggestive of the fact that the 12 pentagons on a fullerene skeleton should be isolated as far as possible due to the graph segments in Figure 1 which primarily contribute to the 8th and 12th moments respectively. Furthermore, the conjugated circuits theory³⁹ also predicts that eight-membered rings destabilize the structure. This is an independent reasoning as to why higher FCIPs are more stable than fullerenes which contain connected or fused pentagons. It turns out that the buckminsterfullerene is the smallest fullerene cage which contains isolated pentagons. Consequently we focus on higher FCIPs in the present paper. According to Euler's rule there should be exactly 12 pentagons for all fullerene cages. It is easily seen that the smallest one of such FCIPs is C_{60} with I_h symmetry. Perhaps this is one of the reasons for the magical stability of C_{60} . In the following, we will derive the exact analytical formulas for the first 14 moments and the coefficients in the characteristic polynomials by the fragmentation method.

Evaluation of Moments of Fullerenes Containing Isolated Pentagons

As is known, the spectral moment of a graph or simply the moment of a graph is an important graph invariant in chemical graph theory. It also connects properties and structures of molecules as well as solid-state materials.²⁰⁻³⁷ Several approaches have been developed to evaluate the moments of graphs derived from molecules to solids. Although the powers of adjacency matrices and the symmetric reduction method facilitate evaluation of moments, they neither reveal the regularity in the moments of specific classes of graphs nor facilitate analytical expressions for a class of graphs. Jiang et al. found an essential relationship between the moments and graph segments.³³ They have expounded this further to a fragmentation approach for evaluating the moments and exploring the relationship between the stability and the reactivities of a molecule with its fragments. Although the fragmentation method is used to evaluate universally the lower level moments (up to μ_{12}), its practical value has been already exhibited via a series of intuitive topological rules for interpreting and predicting the stabilities and reactivities of molecules.

Intuitively the l th moment, μ_l , can be described as the sum of all self-returning walks with l steps over all the edges of a given graph. According to the fragmentation theory, the l th moment is well expressed in an additive form in terms of acyclic moments

* Abstract published in *Advance ACS Abstracts*, September 15, 1993.

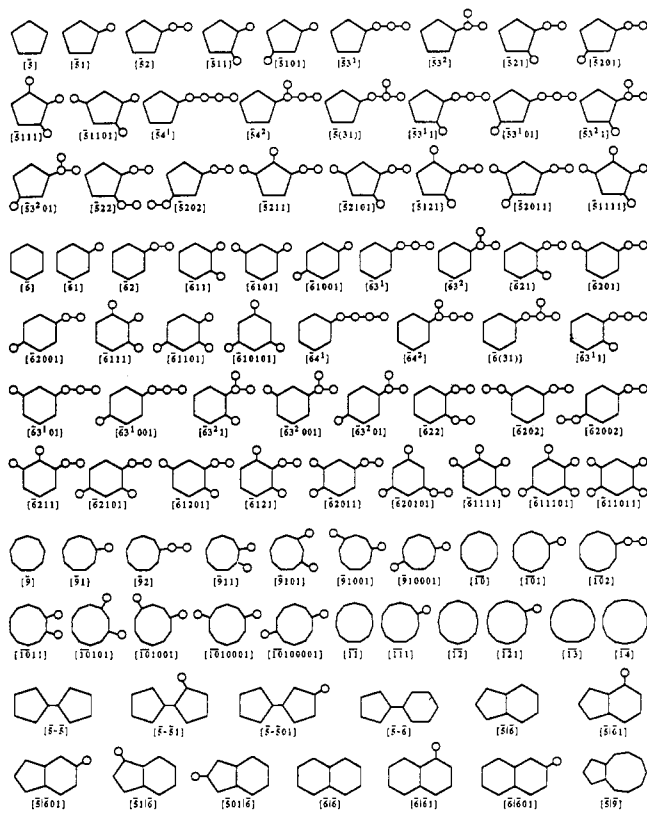


Figure 2. Fragments for cyclic moments of fullerenes.

μ'_l and cyclic moments μ''_l , i.e.

$$\mu_l = \mu'_l + \mu''_l \quad (1)$$

where μ'_l and μ''_l are spanned by acyclic (no loop) and cyclic fragments, G' and G'' , respectively, where "spanned" means the self-returning walk passing through all the edges of the fragments. Furthermore, μ'_l and μ''_l can be expressed as³⁴

$$\mu'_l = \sum_{G'} C_l^{G'} [G'] \quad (2)$$

$$\mu''_l = \sum_{G''} C_l^{G''} [G''] \quad (3)$$

where $C_l^{G'}$ ($C_l^{G''}$) enumerates the number of self-returning walks of length l spanned by the fragments G' (G''), and $[G']$ ($[G'']$) represents the number of fragment G' , (G'').

It should be noted that the acyclic moment μ'_l vanishes when l is odd, which facilitates expressing eq 2 as

$$\mu'_{2l} = \sum_{G'} C_l^{G'} [G'] \quad \mu'_{2l+1} = 0 \quad (4)$$

Jiang and Zhang³³⁻³⁶ have developed eqs 2 and 3 up to μ'_{14} and μ'_{12} for alternant graphs. Since fullerenes are not alternants as they contain 12 pentagons, it is necessary to extend the fragmentation approach for fullerene graphs.

As there are no differences between alternants and nonalternants in acyclic moments μ'_{2l} , we need to consider only the cyclic moments μ''_{2l} . Here we will use the same symbols as in ref 34 to label the generated fragments. Figure 2 shows all the fragments required to evaluate the moments μ''_l up to $l = 14$.

By collecting the coefficients of $C_l^{G''}$ for the fragments in Figure 1 the cyclic moments μ''_l can be obtained explicitly via the following formulas for FCIPs:

$$\mu''_5 = 10[\bar{5}]$$

$$\mu''_6 = 12[\bar{6}]$$

$$\mu''_7 = 70[\bar{5}] + 14[\bar{5}1]$$

$$\mu''_8 = 96[\bar{6}] + 16[\bar{6}1]$$

$$\mu''_9 = 360[\bar{5}] + 144[\bar{5}1] + 18[\bar{5}2] + 18[\bar{5}11] + 18[\bar{5}101] + 18[\bar{9}]$$

$$\mu''_{10} = 540[\bar{6}] + 180[\bar{6}1] + 20[\bar{6}2] + 20[\bar{6}11] + 20[\bar{6}101] + 20[\bar{6}1001] + 20[\bar{10}] + 60[\bar{5}]$$

$$\mu''_{11} = 1650[\bar{5}] + 1012[\bar{5}1] + 220[\bar{5}2] + 264[\bar{5}11] + 242[\bar{5}101] + 198[\bar{9}] + 22[\bar{5}3^1] + 44[\bar{5}3^2] + 22[\bar{5}21] + 22[\bar{5}201] + 22[\bar{5}111] + 22[\bar{5}1101] + 22[\bar{9}1] + 22[\bar{11}] + 66[\bar{5}\bar{6}]$$

$$\mu''_{12} = 2724[\bar{6}] + 1344[\bar{6}1] + 264[\bar{6}2] + 312[\bar{5}11] + 288[\bar{6}101] + 288[\bar{6}1001] + 660[\bar{5}] + 240[\bar{10}] + 24[\bar{6}3^1] + 48[\bar{6}3^2] + 24[\bar{6}21] + 24[\bar{6}201] + 24[\bar{6}2001] + 24[\bar{6}111] + 24[\bar{6}1101] + 24[\bar{6}10101] + 72[\bar{6}\bar{6}] + 144[\bar{5}1] + 48[\bar{5}-\bar{5}] + 24[\bar{10}] + 24[\bar{12}]$$

$$\mu''_{13} = 7150[\bar{5}] + 6058[\bar{5}1] + 1768[\bar{5}2] + 2496[\bar{5}11] + 2132[\bar{5}101] + 1404[\bar{9}] + 312[\bar{5}3^1] + 676[\bar{5}3^2] + 364[\bar{5}21] + 338[\bar{5}201] + 416[\bar{5}111] + 390[\bar{5}1101] + 312[\bar{9}1] + 286[\bar{11}] + 1274[\bar{5}\bar{6}] + 26[\bar{5}4^1] + 52[\bar{5}4^2] + 52[\bar{5}3^201] + 26[\bar{5}22] + 26[\bar{5}202] + 26[\bar{5}211] + 26[\bar{5}2101] + 26[\bar{5}21001] + 26[\bar{5}2011] + 26[\bar{5}1111] + 26[\bar{9}2] + 26[\bar{9}11] + 26[\bar{9}101] + 26[\bar{9}1001] + 26[\bar{9}10001] + 26[\bar{11}1] + 26[\bar{13}] + 78[\bar{5}\bar{6}1] + 78[\bar{5}1\bar{6}01] + 78[\bar{5}1\bar{6}] + 78[\bar{5}01\bar{6}]$$

$$\mu''_{14} = 13104[\bar{6}] + 8596[\bar{6}1] + 2240[\bar{6}2] + 3080[\bar{6}11] + 2660[\bar{6}101] + 2632[\bar{6}1001] + 1820[\bar{10}] + 4620[\bar{5}] + 364[\bar{6}3^1] + 728[\bar{6}3^2] + 448[\bar{6}21] + 420[\bar{6}201] + 392[\bar{6}2001] + 1960[\bar{6}111] + 994[\bar{6}1101] + 504[\bar{6}10101] + 1596[\bar{6}1\bar{6}] + 364[\bar{10}1] + 336[\bar{12}] + 2100[\bar{5}1] + 1680[\bar{5}-\bar{5}] + 28[\bar{6}4^1] + 56[\bar{6}4^2] + 56[\bar{6}(31)] + 28[\bar{6}3^11] + 28[\bar{6}3^101] + 28[\bar{6}3^1001] + 56[\bar{6}3^2] + 56[\bar{6}3^201] + 56[\bar{6}3^2001] + 28[\bar{6}202] + 28[\bar{6}2002] + 28[\bar{6}211] + 28[\bar{6}2101] + 28[\bar{6}21001] + 28[\bar{6}21001] + 28[\bar{6}201] + 28[\bar{6}20101] + 28[\bar{6}1111] + 28[\bar{6}11101] + 28[\bar{6}11011] + 84[\bar{6}\bar{6}1] + 84[\bar{6}1\bar{6}01] + 84[\bar{5}\bar{9}] + 28[\bar{10}2] + 28[\bar{10}11] + 28[\bar{10}101] + 28[\bar{10}1001] + 28[\bar{10}10001] + 28[\bar{12}1] + 28[\bar{14}] + 196[\bar{5}2] + 196[\bar{5}11] + 196[\bar{5}101] + 56[\bar{5}-\bar{5}1] + 56[\bar{5}-\bar{5}01] + \dots \quad (5)$$

Although in general we need a large number of fragments to evaluate the moments, they can be greatly reduced for specific systems. In the case of FCIPs only six variables shown below are required to generate the moments up to μ_{14} :

n : the number of carbon atoms

p : the number of bonds connecting two pentagons as in Figure 3

q : the number of hexagons lying between two pentagons as in Figure 3

r : the number of groups of three hexagons fused in a linear manner (Figure 3)

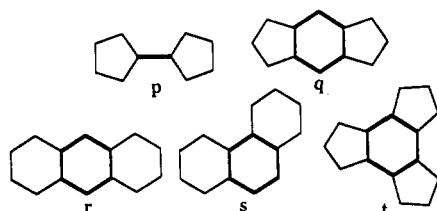


Figure 3. Structural parameters and features for fullerenes.

s: the number of phenanthrene types of fused hexagons as in Figure 3

r: the number of hexagons fused with pentagons on three of the sides as in Figure 3

We can express the fragments contained in eq 19 in ref 33 and eq 5 in terms of *n*, *p*, *q*, *r*, *s*, and *t* for FCIPs. For acyclic fragments, we get

$$\begin{array}{lll}
 [2] = \frac{3}{2}n & [3] = 3n & [4] = 6n \\
 [31] = n & [5] = 12n & [41] = 6n \\
 [6] = 24n - 60 & [51] = 12n & [5101] = 12n \\
 [511] = \frac{3}{2}n & [7] = 42n - 300 & [61] = 24n - 120 \\
 [601] = 48n - 120 & [611] = 12n & [610] = 3n \\
 [502] = 8n & &
 \end{array} \quad \dots (6a)$$

For cyclic fragments we get

$$\begin{array}{lll}
 [\bar{3}] = 12 & [\bar{3}1] = 60 & [\bar{3}2] = 120 \\
 [\bar{3}11] = 60 & [\bar{3}101] = 60 & [\bar{3}3^1] = 240 \\
 [\bar{3}3^2] = 60 & [\bar{3}21] = 240 & [\bar{3}201] = 240 \\
 [\bar{3}111] = 60 & [\bar{3}1001] = 60 & [\bar{3}-\bar{3}] = p \\
 [\bar{6}] = \frac{1}{2}(n-20) & [\bar{6}1] = 3(n-20) & [\bar{6}2] = 6(n-20) \\
 [\bar{6}11] = 3(n-20) & [\bar{6}101] = 3(n-20) & [\bar{6}1001] = \frac{3}{2}(n-20) \\
 [\bar{6}3^1] = 12(n-20) & [\bar{6}3^2] = 3(n-20) & [\bar{6}21] = 12(n-20) \\
 [\bar{6}201] = 12(n-20) & [\bar{6}2001] = 6(n-20) & [\bar{6}111] = n-20 \\
 [\bar{6}1101] = 6(n-20) & [\bar{6}10101] = n-20 & [\bar{5}1\bar{6}] = 60 \\
 [\bar{6}1\bar{6}] = \frac{3}{2}n - 60 & [\bar{9}] = 60 & [\bar{9}1] = 420 \\
 [\bar{10}] = \frac{3}{2}n - 60 & [\bar{10}] = 12n - 480 & [\bar{12}] = 5p + q
 \end{array} \quad \dots (6b)$$

Substituting eqs 6a and 6b into eq 19 in ref 33 and eq 5, we can express the moments up to μ_{14} in terms of independent variables *n*, *p*, *q*, *r*, *s*, and *t* for FCIPs. Notice that the coefficient of $N_{[502]}$ in μ_{14}^G of eq 19 in ref 33 should be 336 instead of 366:

$$\begin{aligned}
 \mu_2 &= 3n \\
 \mu_4 &= 15n \\
 \mu_5 &= 120 \\
 \mu_6 &= 93n - 120 \\
 \mu_7 &= 1680 \\
 \mu_8 &= 639n - 1920 \\
 \mu_9 &= 18360 \\
 \mu_{10} &= 4653n - 22680 \\
 \mu_{11} &= 184800 \\
 \mu_{12} &= 35169n + 120p + 24q - 240120 \\
 \mu_{13} &= 1790880 - 260p - 52q \\
 \mu_{14} &= 264204n - 882q + 1518r + 1918s - 1893780 \quad (7)
 \end{aligned}$$

It is interesting that up to μ_{11} , the even amounts are linear in *n*, the number of vertices, while the odd moments are simply

constants. This means that the isomers of FCIPs should have the same moments up to μ_{11} . Also, the odd moments up to and including μ_{11} are constants.

Analytical Formulas of the First 15 Coefficients of the Characteristic Polynomials of FCIPs

For a given graph *G* with *N* vertices, the characteristic polynomials, $P_G(x)$, is defined as follows:

$$P_G(x) = |Ix - A| = x^N + a_1x^{N-1} + a_2x^{N-2} + \dots + a_N \quad (8)$$

where *I* is the identity matrix, *A* is the adjacency matrix of the graph *G*, and a_1, a_2, \dots, a_N are the 1st, 2nd, ..., *N*th coefficients of $P_G(x)$. It is well-known that the coefficients in eq 8 are one of the important graph invariants and very meaningful in chemical applications.²⁴⁻²⁹ Following the development of Sach's theorem, several methods have been proposed to calculate the characteristic polynomials.²⁴⁻²⁹ The moment method, especially to compute the exact analytical formulas for the first 14 coefficients of the characteristic polynomials, is emphasized here. Jiang and Tang³⁵ derived the following relation between the moments and the coefficients of the characteristic polynomials:

$$a_k = \sum_{(m_1, m_2, \dots, m_k)} \prod_{l=1}^k \frac{1}{m_l!} \left(-\frac{\mu_l}{l}\right)^{m_l} \quad (9)$$

where (m_1, m_2, \dots, m_k) represents a numerical set of *k* integers such that

$$\sum_{l=1}^k l m_l = k \quad (10)$$

Equation 9 has been successfully used for bipartite graphs.^{34,35} Of course, it is a good formula to evaluate the coefficient, a_k , for fullerenes in case that the moments have been calculated.

In eq 9, the summation runs over all the possible ways to construct (m_1, m_2, \dots, m_k) according to eq 10 which should equal the number of partitions of integer *k*, $P(k)$, or number of irreducible representations of permutation group S_k . For convenience, one usually uses a code, $1^{m_1}2^{m_2}\dots k^{m_k}$, to distinguish the distinct selections of m_1, m_2, \dots, m_k that satisfy eq 10 which corresponds to a multiplication term in eq 8. For example, when $k = 4$, the five possibilities $1 \times 4 = 4$, $1 \times 2 + 2 \times 1 = 4$, $1 \times 1 + 3 \times 1 = 4$, $2 \times 2 = 4$ and $4 \times 1 = 4$ are represented by the partitions of the integer 4 which can be represented by the Young diagrams for 1^4 , $1^2 \cdot 2$, $1 \cdot 3$, 2^2 , and 4, respectively. Accordingly

$$\begin{aligned}
 a_4 = \frac{1}{4!} \left(-\frac{\mu_1}{1}\right)^4 &+ \frac{1}{2!} \left(-\frac{\mu_1}{1}\right)^2 \left(-\frac{\mu_2}{2}\right)^2 &+ \left(-\frac{\mu_1}{1}\right) \left(-\frac{\mu_3}{3}\right) &+ \frac{1}{2!} \left(-\frac{\mu_2}{2}\right)^2 &+ \left(-\frac{\mu_4}{4}\right) \\
 \uparrow &\uparrow &\uparrow &\uparrow &\uparrow \\
 1^4 &1^2 \cdot 2 &1 \cdot 3 &2^2 &4
 \end{aligned} \quad (11)$$

Notice that μ_1 and μ_3 are always zero for all fullerenes, so that any multiplication term containing μ_1 and μ_3 in eq 9 should vanish. Consequently, eqs 9 and 10 are simplified as

$$a_k = \sum_{(m_2, m_4, m_5, \dots, m_k)} \prod_{l=1}^k \frac{1}{m_l!} \left(-\frac{\mu_l}{l}\right)^{m_l} \quad (12)$$

$$2m_2 + 4m_4 + 5m_5 + \dots + km_k = k \quad (13)$$

corresponding to eqs 12 and 13, the code $1^{m_1}2^{m_2}\dots k^{m_k}$ is changed into $2^{m_2}4^{m_4}5^{m_5}\dots k^{m_k}$. Table I shows all the codes for $k = 2-14$

respectively. Combining Table I and eq 12, we can get the explicit formulas of a_k ($k = 2-14$) in terms of moments for FCIPs as follows:

$$\begin{aligned}
 a_2 &= \left(-\frac{\mu_2}{2}\right) \\
 a_4 &= \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2 + \left(-\frac{\mu_4}{4}\right) \\
 a_5 &= \left(-\frac{\mu_5}{5}\right) \\
 a_6 &= \frac{1}{3!}\left(-\frac{\mu_2}{2}\right)^3 + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_4}{4}\right) + \left(-\frac{\mu_6}{6}\right) \\
 a_7 &= \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_5}{5}\right) + \left(-\frac{\mu_7}{7}\right) \\
 a_8 &= \frac{1}{4!}\left(-\frac{\mu_2}{2}\right)^4 + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_4}{4}\right) + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_6}{6}\right) + \\
 &\quad \frac{1}{2!}\left(-\frac{\mu_4}{4}\right)^2 + \left(-\frac{\mu_8}{8}\right) \\
 a_9 &= \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_5}{5}\right) + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_7}{7}\right) + \\
 &\quad \left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_5}{5}\right) + \left(-\frac{\mu_9}{9}\right) \\
 a_{10} &= \frac{1}{5!}\left(-\frac{\mu_2}{2}\right)^5 + \left(-\frac{\mu_2}{2}\right)^3\left(-\frac{\mu_4}{4}\right) + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_6}{6}\right) + \\
 &\quad \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_4}{4}\right)^2 + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_8}{8}\right) + \left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_6}{6}\right) + \\
 &\quad \frac{1}{2!}\left(-\frac{\mu_5}{5}\right)^2 + \left(-\frac{\mu_{10}}{10}\right) \\
 a_{11} &= \frac{1}{3!}\left(-\frac{\mu_2}{2}\right)^3\left(-\frac{\mu_5}{5}\right) + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_7}{7}\right) + \\
 &\quad \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_5}{5}\right) + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_9}{9}\right) + \\
 &\quad \left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_7}{7}\right) + \left(-\frac{\mu_5}{5}\right)\left(-\frac{\mu_6}{6}\right) + \left(-\frac{\mu_{11}}{11}\right) \\
 a_{12} &= \frac{1}{6!}\left(-\frac{\mu_2}{2}\right)^6 + \frac{1}{4!}\left(-\frac{\mu_2}{2}\right)^4\left(-\frac{\mu_4}{4}\right) + \\
 &\quad \frac{1}{3!}\left(-\frac{\mu_2}{2}\right)^3\left(-\frac{\mu_6}{6}\right) + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_4}{4}\right)^2 + \\
 &\quad \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_8}{8}\right) + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_6}{6}\right) + \\
 &\quad \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_5}{5}\right)^2 + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_{10}}{10}\right) + \frac{1}{3!}\left(-\frac{\mu_4}{4}\right)^3 + \\
 &\quad \left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_8}{8}\right) + \left(-\frac{\mu_5}{5}\right)\left(-\frac{\mu_7}{7}\right) + \frac{1}{2!}\left(-\frac{\mu_6}{6}\right)^2 + \left(-\frac{\mu_{12}}{12}\right) \\
 a_{13} &= \frac{1}{4!}\left(-\frac{\mu_2}{2}\right)^4\left(-\frac{\mu_5}{5}\right) + \frac{1}{3!}\left(-\frac{\mu_2}{2}\right)^3\left(-\frac{\mu_7}{7}\right) + \\
 &\quad \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_5}{5}\right) + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_9}{9}\right) + \\
 &\quad \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_7}{7}\right) + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_5}{5}\right)\left(-\frac{\mu_6}{6}\right) + \\
 &\quad \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_{11}}{11}\right) + \frac{1}{2!}\left(-\frac{\mu_4}{4}\right)^2\left(-\frac{\mu_5}{5}\right) + \left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_9}{9}\right) + \\
 &\quad \left(-\frac{\mu_5}{5}\right)\left(-\frac{\mu_8}{8}\right) + \left(-\frac{\mu_6}{6}\right)\left(-\frac{\mu_7}{7}\right) + \left(-\frac{\mu_{13}}{13}\right)
 \end{aligned}$$

$$\begin{aligned}
 a_{14} &= \frac{1}{7!}\left(-\frac{\mu_2}{2}\right)^7 + \frac{1}{5!}\left(-\frac{\mu_2}{2}\right)^5\left(-\frac{\mu_4}{4}\right) + \\
 &\quad \frac{1}{4!}\left(-\frac{\mu_2}{2}\right)^4\left(-\frac{\mu_6}{6}\right) + \frac{1}{3!2!}\left(-\frac{\mu_2}{2}\right)^3\left(-\frac{\mu_4}{4}\right)^2 + \\
 &\quad \frac{1}{3!}\left(-\frac{\mu_2}{2}\right)^3\left(-\frac{\mu_8}{8}\right) + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_6}{6}\right) + \\
 &\quad \frac{1}{2!2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_5}{5}\right)^2 + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)^2\left(-\frac{\mu_{10}}{10}\right) + \\
 &\quad \frac{1}{3!}\left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_4}{4}\right)^3 + \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_8}{8}\right) + \\
 &\quad \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_5}{5}\right)\left(-\frac{\mu_7}{7}\right) + \frac{1}{2!}\left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_6}{6}\right)^2 + \\
 &\quad \left(-\frac{\mu_2}{2}\right)\left(-\frac{\mu_{12}}{12}\right) + \frac{1}{2!}\left(-\frac{\mu_4}{4}\right)^2\left(-\frac{\mu_6}{6}\right) + \\
 &\quad \frac{1}{2!}\left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_5}{5}\right)^2 + \left(-\frac{\mu_4}{4}\right)\left(-\frac{\mu_{10}}{10}\right) + \left(-\frac{\mu_5}{5}\right)\left(-\frac{\mu_9}{9}\right) + \\
 &\quad \left(-\frac{\mu_6}{6}\right)\left(-\frac{\mu_8}{8}\right) + \frac{1}{2!}\left(-\frac{\mu_7}{7}\right)^2 + \left(-\frac{\mu_{14}}{14}\right) \quad (14)
 \end{aligned}$$

Substituting eq 7 into eq 12, the following expressions for the first 14 coefficients of the characteristic polynomials of fullerenes, namely, a_k ($k \leq 14$) are obtained as a function of n , p , q , r , and s (note that a_1 and a_3 are zeroes):

$$\begin{aligned}
 a_2 &= -^3/2n \\
 a_4 &= ^1/8(9n^2 - 30n) \\
 a_5 &= -24 \\
 a_6 &= -^1/16(9n^3 - 90n^2 + 248n - 320) \\
 a_7 &= 36n - 240 \\
 a_8 &= ^1/128(27n^4 - 540n^3 + 3876n^2 - 14064n) + 240 \\
 a_9 &= -27n^2 + 450n - 2040 \\
 a_{10} &= -^1/1280(81n^5 - 2700n^4 + 35820n^3 - 256560n^2 + \\
 &\quad 1152384n) + 2556 \\
 a_{11} &= ^1/2(27n^3 - 810n^2 + 8664n) - 17280 \\
 a_{12} &= ^1/5120(81n^6 - 4050n^5 + 85140n^4 - 1009080n^3 + \\
 &\quad 7680544n^2 - 40830720n) + 25970 - 10p - 2q \\
 a_{13} &= -^1/16(81n^4 - 3780n^3 + 69948n^2 - 627312n) - \\
 &\quad 148320 + 20p + 4q \\
 a_{14} &= -^1/71680(243n^7 - 17010n^6 + 517860n^5 - 9155160n^4 + \\
 &\quad 146500032n^3 - 875226240n^2 + 5213230080n) + 217630 + \\
 &\quad 15np + 3nq + 63q - 137r - 137s \quad (15)
 \end{aligned}$$

The above expressions for the coefficients of the characteristic polynomials are also interesting results in number theory. For example, from the expression for a_{12} we infer that for any even integer $n \geq 60$, $81n^6 - 4050n^5 + 85140n^4 - 1009080n^3 + 7680544n^2 - 40830720n$ is divisible by 5120. And we can infer from a_{10} that $81n^5 - 2700n^4 + 35820n^3 - 256560n^2 + 1152384n$ is divisible by 1280 for any even integer $n \geq 20$.

It can be seen from eq 15 that for $k \leq 11$, the coefficient, a_k , is structure independent in that the coefficient depends only on n (the number of carbon atoms) for FCIPs. This can be attributed to the fact that for $k \leq 11$, the moment, μ_k , can be expressed as a linear form of n for FCIPs via eqs 8 and 9. For a_k ($k \geq 12$) the topological differences between the isomers are exhibited in the characteristic polynomials, which would determine the differences among the isomers of FCIPs in some properties, such as the topological stability.

TABLE I: Codes for $k = 2-14$

k	$\{2m_2, 4m_4, 5m_5, \dots, km_k\}$
2	2
4	2 ² , 4
5	5
6	2 ³ , 2·4, 6
7	2·5, 7
8	2 ⁴ , 2 ² ·4, 2·6, 4 ² , 8
9	2 ² ·5, 2·7, 4·5, 9
10	2 ⁵ , 2 ³ ·4, 2 ² ·6, 2·8, 4·6, 5 ² , 10
11	2 ³ ·5, 2 ² ·7, 2·4·5, 2·9, 4·7, 5·6, 11
12	2 ⁶ , 2 ⁴ ·4, 2 ³ ·6, 2 ² ·4 ² , 2 ² ·8, 2·4·6, 2·5 ² , 2·10, 4 ³ , 4·8, 5·7, 6 ² , 12
13	2 ⁴ ·5, 2 ³ ·7, 2 ² ·4·5, 2 ² ·9, 2·4·7, 2·5·6, 2·11, 4 ² ·5, 4·9, 5·8, 6·7, 13
14	2 ⁷ , 2 ⁵ ·4, 2 ⁴ ·6, 2 ³ ·4 ² , 2 ³ ·8, 2 ² ·4·6, 2 ² ·5 ² , 2 ² ·10, 2·4 ³ , 2·4·8, 2·5·7, 2·6 ² , 2·12, 4 ² ·6, 4·5 ² , 4·10, 5·9, 6·8, 7 ² , 14

It is well-known that the total π -electron energy can be expressed in an additive form in terms of the moments, the Legendre polynomial, and Chebyshev's polynomial. Parameter-fitting methods have also been used to develop expressions for these quantities. A general regularity can be obtained from the expression that the $(4n + 2)$ th moments lower the total energy while the $4n$ th moments increase the total energy. The contribution of moments to the energy decreases greatly as the order of moments increases. Similar regularity is found for molecular fragments, which is called the generalized Hückel rule.³⁴ According to the proposed structure of C_{60} , large fullerene cages are typically π -electron systems, to which the above-mentioned rules could be applied. Therefore, the relative stability among isomers of FCIPs can be determined by the moments although the reliability of the prediction is no more than the Hückel method of π -electronic systems. However, the moment method provides significant insight into the question of which features in a structure enhance stability and which features would destabilize structure. That is, it is uniquely equipped to identify the topological features which result in the enhancement of stabilities and features which are undesirable in the structure. Since there are no differences in the 11th or lower moments, the following rules could be implemented.

(1) The structure with larger μ_{12} would be less stable, where

$$\Delta\mu_{12} = 120\Delta p + 24\Delta q \quad (16)$$

(2) For isomers with the same μ_{12} , the one with smaller μ_{14} would be less stable:

$$\Delta\mu_{14} = -882\Delta q + 1918(\Delta r + \Delta s) \quad (17)$$

(3) For the ones with the same μ_{12} and μ_{14} , one could continue to analyze μ_{16} and so on.

C_{78} is the simplest example to illustrate the procedure. There are five FCIP isomers for C_{78} among which two have D_{3h} , one has D_3 and the other two have C_{2v} symmetries as shown in Figure 4. By counting variables p and q and using eq 16, we can get the trends of stability:

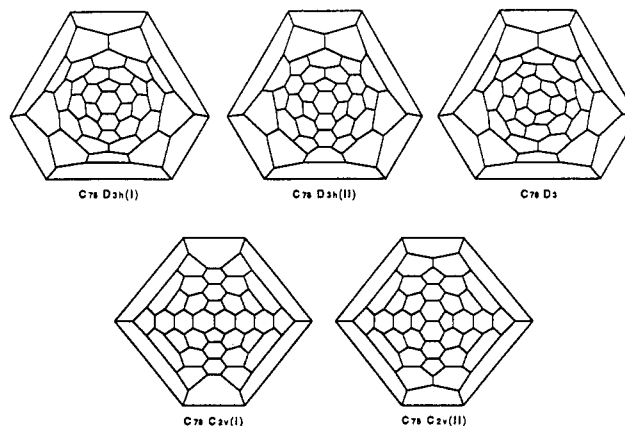
$$D_{3h}(I) < D_3 < C_{2v}(I) < C_{2v}(II) < D_{3h}(II)$$

Although we do not expect the above prediction to be vigorously valid at higher levels of theory, the method is a good starting point to select a few alternative candidates for higher order computations.

Finally, it should be emphasized that the moments and characteristic polynomials are equivalent graph invariants, since the following relation³⁵ holds:

$$-ka_k = \mu_1 a_{k-1} + \mu_2 a_{k-2} + \dots + \mu_{k-1} a_1 + \mu_k \quad (18)$$

Therefore, to determine the relative stability by moments is tantamount to using the coefficients of the characteristic polynomials.

Figure 4. Schlegel diagrams of five C_{78} isomers.

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