



## How to Have Been Struggling with the Beauty and Symmetry of Soccer Ball Fullerene–Personal History

Haruo Hosoya \*

Ochanomizu University (Emeritus), Bunkyo-ku, Tokyo 112-8610, Japan

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On this occasion I thought that it is meaningful to trace back and document my personal history involved in this beautiful soccer ball shape and molecule  $C_{60}$  not only for myself but also for the next generation to follow. Therefore, the topics may be moving to and fro in the 4-dimensional world. If the readers find any inaccurate description, please, remind its corrections or additions to me privately or to the public freely.

**1936 = 44<sup>2</sup>**

I was born in Kamakura, an old capital town, 50km south of Tokyo, Japan, and grew up there until I flew with my wife and first baby son to North America in 1967. In Montreal I visited EXPO and saw Buckminster-Fuller Dome of US pavilion. However, at that time I didn't get special impact from that. Then I went to Ann Arbor, Michigan, to do post-doctoral work under the guidance of John Platt from Chicago. Connection between us was made in 1962 when he came to Japan to attend International Symposium on Molecular Structure and Spectroscopy, and after this big academic event I took him with several of my friends to the top of Mt. Fuji, 3776m above sea level. During one year of stay in Michigan I was a biophysicist struggling with the eye-balls of rats for vision research.

After this unusual experience, in April of 1969, I became associate professor of chemistry in Ochanomizu University. Among almost 100 national universities in Japan only Ochanomizu University and Nara Women's University are only for women students. Although the fame of my university and the level of the students were very high, the research envi-

\*Corresponding author (Email address: [hosoya.haruo@ocha.ac.jp](mailto:hosoya.haruo@ocha.ac.jp)).

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ronment was so miserable that I had to do everything without research assistant and with nominal amount of research fund, and I was asked to guide three undergraduate students for helping their graduation thesis. Then from April of 1970 two master course students, Keiko Kawasaki and Kimiko Mizutani, began to study in my laboratory, to whom I am very grateful for their effort to build up the foundation of Hosoya laboratory.

In this August I could take Shinkansen, a new bullet train, to visit Osaka EXPO, where again Buckminster-Fuller Dome was waiting for me as the US pavilion, but also in this time I felt no special interest to it.

Alas. Just at that time in Kyoto, only 40km north-east of Osaka EXPO place, Eiji Osawa was writing a breathtaking commentary article predicting the "super-aromatic" soccer ball-shaped  $C_{60}$ . [36,38] I also didn't know that from this year in Mexico truncated eicosahedron ball began to be used officially in the FIFA world cup of soccer.

In the end of this year I could write a chemistry monograph book "Structure and Properties" [35] coauthored by my former boss, Saburo Nagakura, who later became president of IUPAC (International Union of Pure and Applied Chemistry) and was given the highest order from the Government of Japan. However, in science I could learn almost nothing from him.

## Topological Index

Right after changing my research field, in 1971 I published my first graph-theoretical paper, "Topological index. A newly proposed quantity characterizing the topological nature of structural isomers of saturated hydrocarbons" [14]. According to Balaban [5] Hosoya is the "Godfather" and inventor of topological index, and thus my index is now called either Z-index or Hosoya-index.

Define the non-adjacent number,  $p(G,k)$ , as the number of ways for choosing  $k$  disjoint edges from  $G$ , a non-directed simple graph extracted from the carbon atom skeleton of a hydrocarbon molecule. The Z-index of  $G$  composed of  $N$  vertices is the total sum of its  $p(G,k)$  number as

$$Z = \sum_{k=0}^{\lfloor \frac{N}{2} \rfloor} p(G,k). \quad (1)$$

In other words  $Z$  is the total number of matchings of  $G$  plus one. If one defines the Z-counting polynomial by using the set of  $p(G,k)$  numbers as

$$Q_G(x) = \sum_{k=0}^{\lfloor \frac{N}{2} \rfloor} p(G,k)x^k, \quad (2)$$

we have

$$Z = Q_G(1). \quad (3)$$

The characteristic polynomial  $P_G(x)$  of tree graph  $G$  is shown to be obtained by using  $p(G,k)$ 's as

$$P_G(x) = \sum_{k=0}^{\lfloor \frac{N}{2} \rfloor} (-1)^k p(G,k) x^{N-2k} \quad (G \in \text{Tree}), \quad (4)$$

where  $P_G(x)$  is defined in terms of the  $N \times N$  adjacency matrix  $A$  and unit matrix  $E$  as

$$P_G(x) = (-1)^N \det(A - xE). \quad (5)$$

Since for a non-tree graph one needs to add ring correction terms to (4), [16] it is rather difficult to get  $P_G(x)$  of  $C_{60}$  fullerene just from the set of  $p(G,k)$ . In Appendix the  $P_G(x)$  and  $Q_G(x)$  of  $C_{60}$  will be compared.

By piling up the information obtained from  $A$  of  $G$  one can obtain the distance matrix  $D$ , whose element  $D_{ij}$  is the shortest topological path length between vertices  $i$  and  $j$ . In terms of  $D$  we first defined the distance polynomial [15] as

$$S_G(x) = (-1)^N \det(D - xE), \quad (6)$$

in 1973. Since the coefficients of  $S_G(x)$  of  $C_{60}$  are so large, only a part of them is given in Appendix [30].

As the  $P_G(x)$  is directly related to Huckel molecular orbital (HMO) method, my interest was gradually directed to aromatic hydrocarbons and in 1975, we proposed to define the sextet polynomial [17] for a given benzenoid graph  $G$  as

$$B_G(x) = \sum_{k=0}^m r(G,k) x^k, \quad (7)$$

where  $r(G,k)$  is the resonant sextet number, the number of ways in which  $k$  disconnected but resonant sextets are chosen from  $G$  (For details consult the original paper). The  $B_G(x)$  of  $C_{60}$  calculated by Shiu et al. is given in Appendix [37].

## Origami

Aside from research in application of graph theory to chemical and physical problems I got interested in preparing cheap molecular models for students from high school to university. I forgot the year but in some toy shop I found an interesting set of origami (popular square-paper folding in Japan) called "unit (modular) origami". By using many sheets of rectangles with the ratio of  $1 : \sqrt{3}$  a number of different polyhedra can be folded without scissors and glue. It was originally designed by an architect Norishige Terada of Taisei Cooperation, and later I knew that he was in charge of constructing the Fuller-Dome for US pavilion in Osaka EXPO.

Then I got interested in this "unit origami" for designing molecular and crystal models, and finally in 1979 I wrote a series of commentary papers [18–20] in a popular chemistry

journal, Gendai Kagaku (Chemistry Today), "Learn Stereo Chemistry by Using Origami," where a variety of polyhedra are made from origami paper.

See Figure 1 which is a copy of the cover of that journal [19]. Although this is not an origami work, within the cage of a truncated icosahedron doubly overlapped regular icosahedra are embedded. At that time  $C_{60}$  was not discovered, but the structure of a cluster of  $B_{84}(= 60 + 12 + 12)$  was determined precisely, [13] I believe that this is the world-first drawing of truncated icosahedron printed on the cover page of chemical journals and/or books.

On the other hand, in 1981, Davidson [7] succeeded in factoring out the  $60 \times 60$  determinant of  $P_G(x)$  of unknown  $C_{60}$  by repeated use of mirror reflection technique of point-group.

In my laboratory, however, in 1982 I could succeed in making an origami model of truncated icosahedron, which was introduced in my essay published in another popular journal of science (See Figure 2) [21]. It is constructed from 90 sheets of unit origami paper designed by Terada.



Figure 1. Cluster of  $B_{84}$ .



Figure 2. Truncated icosahedron from unit origami.

Thanks to my papers on molecular models with origami a big publisher Iwanami asked to write an introductory book of chemistry, but I hesitated to take an immediate action. Then I tried to use a paper envelope to make many molecular and crystal structures, and in 1983 I could write a book "To Grasp Chemistry" for high school students full of instructions for

making molecular and crystal models by using origami and envelope [22]. This time a soccer ball can be made from an envelope as shown in Figure 3, and on the cover of this book (Figure 4) a paper-made soccer ball is highlighted. For making this model we need precise drawing, scissors, and glue, but a stable polyhedron with holes can be made quite easily. However, still it was not supposed to be  $C_{60}$  but  $B_{84}$  as already explained above. It was two years before the discovery of  $C_{60}$ .

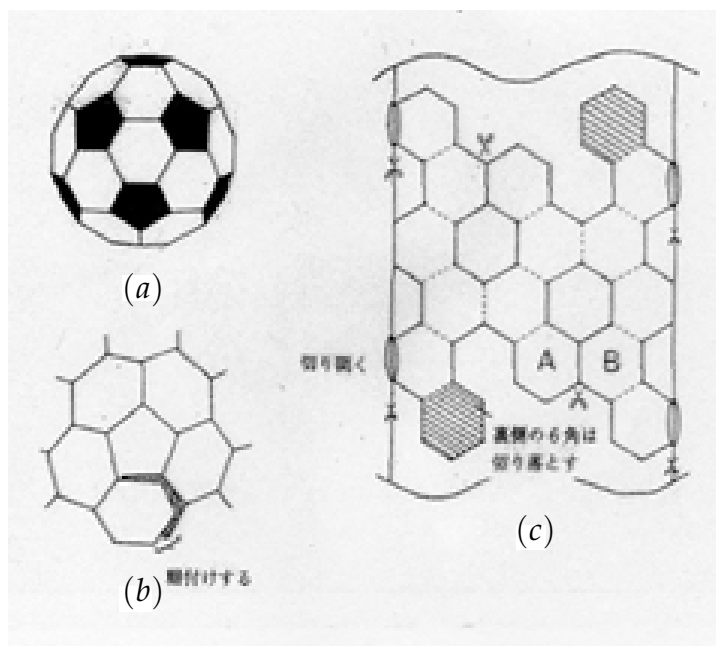


Figure 3. A soccer ball can be made from an envelope.

As seen in Figure 3(c) we drew only regular hexagons but no pentagon at all. However, twelve pentagons are obtained as holes each of which is surrounded by five regular hexagons.

At that time instead of solving  $P_G(x)$  to obtain the energy levels of HMO, I was struggling with the  $p(G,k)$  and  $Z$ -indices of spheroidal polyhedra. Nowadays enumeration of  $p(G,k)$  is known to be a  $NP$ -complex problem, but at that time without knowing such a deep mathematical discussion I was simply struggling with very long machine time for calculating  $p(G,k)$  numbers.

### Mathematical chemists in fullerene science

The University of Tokyo was running IBM 5080 computer, which was the best machine at that time in Japan available to university people. However, the estimated cost for calculating all the coefficients of  $Q_G(x)$  was huge enough for a poor researcher in a small university. Then I decided to get them by decomposing and decomposing the truncated icosahedron



Figure 4. Cover of "To Grasp Chemistry".

into a number of smaller feasible subgraphs by using ad hoc giant recursion formulas. After filling up a whole big notebook I could obtain the final result, but I had no confidence in my result, except for the highly factorable last term, or perfect matching number, of  $12500 = 2^2 \times 5^5$ . What a beautiful result it was. Then I submitted my paper "Matching and Symmetry of Graphs" to Computers and Mathematics with Applications as I was invited by Hargittai to contribute to its special issue [23]. It was published in 1986, and was adopted into a book "Symmetry Unifying Human Understanding" [12].

Fortunately, all the coefficients of my  $Q_G(x)$ , namely, the set of  $p(G,k)$ 's were found to be equal to the matching polynomial (See later) obtained by the group of Klein published in the same year [31]. I was relieved by knowing that my adventurous calculation was correct. These people together with all other chemists had been triggered by the dramatic discovery of  $C_{60}$  by the group of Kroto and Smalley [33] in 1985.

The reason why I was not defeated in this race was simply because I luckily started before other runners gathered to the starting point.

Next in 1987 I wrote a commentary article on the geometrical structure and symmetry of  $C_{60}$  and  $B_{84}$  in a science journal in Japan [24]. There I also explained the important role of the character table of the group theory, which can correctly predict the selection rule of normal vibrations and symmetrical decomposition of molecular orbitals according to the prescribed recipe of point group.

Although it took almost five years for getting experimental evidence for the soccer ball structure of  $C_{60}$ , [32] mathematical chemists quickly established its theoretical support by calculating various quantum chemical quantities for all the possible 1812 isomers of "5,6-

polyhedra". The truncated icosahedron  $C_{60}$  consist of 12 pentagons and 20 hexagons so that 3 faces meet at each of 60 vertices.

However, to establish the isomer number of 1812 a few years of battle was fought among at least three groups of mathematical chemists in US and Europe. Now it is accepted that the "spiral conjecture and algorithm" developed by Fowler and Manolopoulos [8] is practically useful for obtaining the isomer numbers irrespective of its mathematical incompleteness.

Here I would like to point out an important issue on the contribution of mathematical chemists and pure mathematicians in fullerene chemistry. Now a number of mathematicians are working in the field of fullerene science, but as I already mentioned in this paper, the most important part of mathematical findings, as symbolized by the numbers 1812 and 12500, was obtained only by mathematical chemists. The important results obtained by them and relevant discussions are given in several monographs [4,6,8]. Remember that the theory of point group was constructed in 1930's only by physicists and chemists [29]. Now understanding and cooperation from the side of mathematicians are necessary for consolidating the fullerene science.

The perfect matching number of 12500 is the last term of the matching polynomial, which is often used to discuss the aromaticity of polycyclic conjugated hydrocarbons like fullerene.

Let us go back several years for the introduction of matching polynomial, or acyclic polynomial. During 1976-1979 at least three groups of theorists independently proposed the same polynomial

$$M_G(x) = \sum_{k=0}^{\lfloor \frac{N}{2} \rfloor} (-1)^k p(G,k) x^{N-2k}, \quad (8)$$

all by using my  $p(G,k)$  numbers, but with different names. Namely, Aihara, [1] Gutman et al., [11] and Farrell, [10] respectively, call reference, acyclic, and matching polynomials. Recently the most popular name seems to be matching polynomial. The relations between  $M_G(x)$  and  $Q_G(x)$  can easily be obtained as

$$M_G(x) = x^N Q_G(-x^{-2}), \quad (9)$$

and

$$Q_G(x) = (-i\sqrt{x})^N M_G\left(\frac{i}{\sqrt{x}}\right). \quad (10)$$

By using his theory of aromaticity in 1988 Aihara wrote a paper on the spherical aromaticity of  $C_{60}$  by saying that aromatic stabilization of  $C_{60}$  is not large enough to guarantee isolation in macroscopic amounts but this does not rule out the possibility that such carbon clusters are formed in sooting flames and/or in dark interstellar space. He kindly put my name into the author of this paper, [2] and in a Nobel prize-campaign paper by Kroto et al. [34] they cited this paper and  $K = 12500$  papers by me and Klein's group [23,31].

In this year I wrote a paper "On some counting polynomials in chemistry," in which Wiener polynomial was proposed to be defined as

$$H_G(x) = \sum_{k=1}^l d(G,k)x^k, \quad (11)$$

where  $d(G,k)$  is the number of vertex pair with shortest distance  $k$  in  $G$ , and  $l$  is the largest element of  $D$ , [25] but this polynomial is now called Hosoya polynomial. By taking the formal derivative of  $H_G(x)$  with respect to  $x$ , one can obtain the Wiener index  $w$  [14] as follows:

$$w = \sum_{k=1}^l k d(G,k) = H'_G(1), \quad (12)$$

The  $H_G(x)$  and  $w$  calculated for  $C_{60}$  by Arezoomand is given in Appendix [3].

Almost for ten years after the paper by Kroto et al. [33] the fullerene-boom was overwhelming the chemists at large, and finally in 1996 the Nobel Prize of chemistry was given to Kroto, Smalley and Curl, but not to my friend Osawa. After this big event I was asked to write commentary articles and to give a lecture quite often. The favorite title of my talk to high school students and teachers was "Soccer ball in the universe" [28]. However, when I was invited to join some chemical educational workshop held in a suburb of London, I changed my title into "Football in the universe".

### Soccer ball in the universe

The outline of my talk under this title is usually as follows:

- 1) History of the official soccer ball in FIFA, which can be followed and studied by the postal stamps in many countries all over the world
- 2) Geometry of truncated icosahedron, regular and semi-regular polyhedra
- 3) Euler's theorem,  $E = V + F - 2$
- 4) Symmetry of polyhedra and point group
- 5) Discovery of  $C_{60}$  and nanotube
- 6) Chemical importance of fullerene
- 7)  $C_{60}$  in meteorite and nebula

Brief comment to 6) will be given here. By the discovery of fullerene including  $C_{60}$  it was shown that the allotropes of carbon can exist in the world of various dimensions from 3 to 0. Namely, diamond in 3D, graphite or graphene in 2D, carbyne ( $\cdots C-C\equiv C-C\equiv C\cdots$ ) in 1D,



and finally fullerene in 0D. Even a large molecule such as  $C_{60}$ , it is an entity in 0D world, because its size is finite and can be deemed as though zero in comparison with infinitely large carbyne in 1D world. It can be said that Kroto tried to get 1D entity by decomposing 2D entity, but obtained an entity in 0D world.

As remarked in 1) up to now in almost all the countries so many postal stamps have been published. The total number in the world may go up to one thousand. The history of the design of the official ball of FIFA can be traced just by collecting the soccer ball stamps of high quality. For example, Mongolia published a good series of stamps on this matter. However, the culture of Japan in this category was very low until 2002 when the world cup was held jointly in Japan and Korea. I think still in 21<sup>st</sup> century the total number of soccer-relevant stamps published in my country is as few as twenty, and only a few of them can be used as a research material for the history of soccer ball.

Incidentally a funny thing happened to me in this problem. It was in November of 1983. I found a big mistake in Asahi Shimbun, one of the major newspapers in Japan. They were using a small cut of a soccer ball on top of the report of soccer event, but the ball design was wrong. A black hexagon was surrounded by six white hexagons! No pentagon at all. Then I wrote a letter to the editorial board of Asahi pointing out their error, together with a copy of my book, *To Grasp Chemistry*, and waited for a few days, and found that they changed into a new correct drawing of a soccer ball. However, I have never got any answer nor acknowledgment from them. In my letter I wrote that nowadays even in comic journals for kids a soccer ball was correctly drawn, which might have hurt their pride.

### Topological symmetry

In my research of fullerene I was always sticking to the symmetry of the soccer ball,  $C_{60}$  (See Figure 5a). Besides the important number of 12500 [23] I think I have made at least two contributions in this problem. The solution of  $P_G(x) = 0$  of a conjugated hydrocarbon gives the "spectrum" and total  $\pi$ -electronic energy  $E\pi$ . As evident from Figure 5c the orbital energy levels, or the spectrum (solution of  $P_G(x) = 0$ ) of  $C_{60}$ , are highly degenerate much more than other symmetrical graphs appearing in chemistry. Namely, this factorizable feature can be anticipated by handling the character table used in point group manipulation, but much more than expected. Then I tried to rearrange the Schlegel diagram (Figure 5b) into the diagram shown in Figure 5d with ten-fold rotational symmetry in topological sense. Then its  $P_G(x)$  can easily be factored out by using the unit graph as drawn in Figure 5e, which can explain the mathematical structure of Figure 5c. This idea of topological symmetry can be applied to other symmetrical graphs even to regular octahedron [6, 26].

### Kaleidoscope

On the other hand, by using the 3D geometrical symmetry I could design and make a soccer ball Kaleidoscope, the main part of which is composed of five congruent triangular

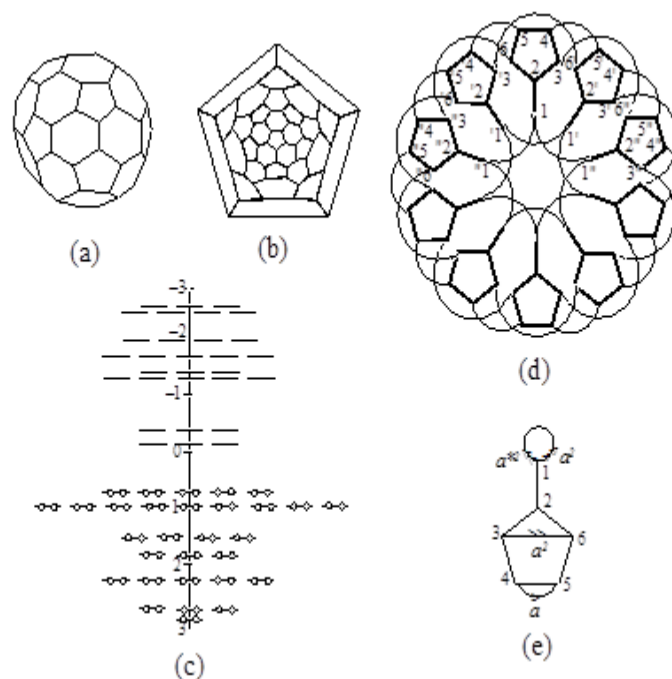


Figure 5. Topological symmetry of  $C_{60}$  [6,26].

mirrors forming a form of pentagonal cone. By peeping into the cone one can only see some complicated reflection pattern of five-fold symmetry. However, if one puts a specially designed small piece properly into the cone, a beautiful soccer ball appears in it. The secret piece is composed of a regular pentagon surrounded by five skewed pentagons. The original idea came from the late Toshiaki Betsumiya, one of my best puzzle friends. Details can be given in my paper [27].

### Soccer ball items

Finally I will introduce only a part of my collection of soccer ball items, all of which, however, cannot be stored in my display shelf. In Figures 6 and 7 are given photographs of the balls made in Japan (temari) and China (multi-layered ball), respectively. The materials of the items in Japan are all soft threads and clothes, while those in China are ivory, beautiful stone, and fragment wood, all precious materials. The geometrical pattern on the spherical surface of a traditional temari in Japan usually contains great circles, but recently patterns of icosahedral symmetry as of soccer ball are getting popular as shown in Figure 6. On the other hand, curiously enough, the patterns on cheaper multi-layered balls in China have icosahedral symmetry, while the expensive ones symbolized in the "magic ball" in National Palace Museum in Taipei have 14 holes of cuboctahedral symmetry.



Figure 6. Modernized temaris in Japan.



Figure 7. Multi-layered balls made in China.

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**Appendix I.**  $Q_G(x)$  and  $P_G(x)$  of soccer ball  $C_{60}$ .

$Q_G(x)$	$P_G(x)$	
1	$x^{60}$	
+90 $x$	$-90 x^{58}$	
+3825 $x^2$	+3825 $x^{56}$	$-24 x^{55}$
+102120 $x^3$	$-102160 x^{54}$	+1920 $x^{53}$
+1922040 $x^4$	+1925160 $x^{52}$	$-72240 x^{51}$
+27130596 $x^5$	$-27244512 x^{50}$	+1700640 $x^{49}$
+298317860 $x^6$	+300906380 $x^{48}$	$-28113600 x^{47}$
+2619980460 $x^7$	$-2661033600 x^{46}$	+347208896 $x^{45}$
+18697786680 $x^8$	+19180834020 $x^{44}$	$-3327625680 x^{43}$
+109742831260 $x^9$	$-114118295000 x^{42}$	+25376437920 $x^{41}$
+534162544380 $x^{10}$	+565407465144 $x^{40}$	$-156652575440 x^{39}$
+2168137517940 $x^{11}$	$-2346799508400 x^{38}$	+792175427520 $x^{37}$
+7362904561730 $x^{12}$	+8189116955350 $x^{36}$	$-3308173115904 x^{35}$
+20949286202160 $x^{13}$	$-24056403184260 x^{34}$	+11466942645600 $x^{33}$
+49924889888850 $x^{14}$	+59443188508110 $x^{32}$	$-33076275953760 x^{31}$
+99463457244844 $x^{15}$	$-123163094844616 x^{30}$	+79417625268960 $x^{29}$
+165074851632300 $x^{16}$	+212712221820840 $x^{28}$	$-158412719276240 x^{27}$
+227043126274260 $x^{17}$	$-303315997028160 x^{26}$	+261359090670624 $x^{25}$
+256967614454320 $x^{18}$	+351861389316780 $x^{24}$	$-354145195147200 x^{23}$
+237135867688980 $x^{19}$	$-324375523213200 x^{22}$	+390055074762240 $x^{21}$
+176345540119296 $x^{20}$	+228227031040884 $x^{20}$	$-344185906596720 x^{19}$
+104113567937140 $x^{21}$	$-122654402736360 x^{18}$	+238553091055200 $x^{17}$
+47883826976580 $x^{22}$	+29617003666920 $x^{16}$	$-126428882536240 x^{15}$
+16742486291340 $x^{23}$	+4679380503120 $x^{14}$	+49433493646080 $x^{13}$
+4310718227685 $x^{24}$	$-8131429397135 x^{12}$	$-13627897407360 x^{11}$
+783047312406 $x^{25}$	+3576552321006 $x^{10}$	+2527365617120 $x^9$
+94541532165 $x^{26}$	$-831616531095 x^8$	$-310065067080 x^7$
+6946574300 $x^{27}$	+108565938200 $x^6$	+26034025632 $x^5$
+269272620 $x^{28}$	$-7440712560 x^4$	$-1566501120 x^3$
+4202760 $x^{29}$	+186416640 $x^2$	+54743040 $x$
+12500 $x^{30}$	+2985984	

$M_G(x)$  can be obtained from  $Q_G(x)$  by using Eq. (9).

**Appendix II.**  $S_G(x)$ ,  $B_G(x)$ ,  $H_G(x)$ , and  $w$  of soccer ball  $C_{60}$ .

$$S_G(x) = x^{60} - 46020x^{58} - 6831720x^{57} \dots + 20689997542195200x + 75501259859424,$$

$$B_G(x) = 1 + 20x + 160x^2 + 660x^3 + 1510x^4 + 1912x^5 + 1240x^6 + 320x^7 + 5x^8,$$

$$H_G(x) = 30(3x + 6x^2 + 8x^3 + 10x^4 + 10x^5 + 10x^6 + 8x^7 + 3x^8 + x^7),$$

$$w = 8340.$$

I'm very glad to have gathered almost all the counting polynomials defined by me (of course, except for  $P_G(x)$ ) for soccer ball  $C_{60}$  here. According to Fowler, the value of  $w$  for the soccer ball is the smallest among 1812 isomers of  $C_{60}$ , quite similar to the case with  $Z$ .