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HARARY–READ NUMBERS FOR CATAFUSENES: COMPLETE CLASSIFICATION ACCORDING TO SYMMETRY

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Abstract

The classical "Harary–Read numbers" for catafusenes (catacondensed simply connected polyhexes) are reproduced without using generating functions. A complete (mathematical) solution is given for the distribution of these numbers over the different symmetry groups to which the catafusenes belong.

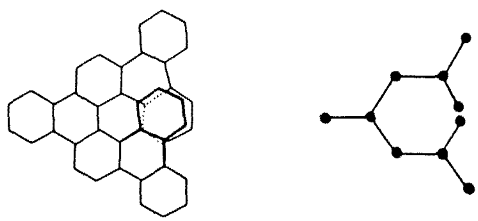
1. Introduction

The classical paper by Harary and Read [1] from 1970 is an outstanding piece of work in the area of polyhex enumeration. The objects which are enumerated therein are the catacondensed simply connected polyhexes, which are referred to as catafusenes [2]. They are without internal vertices and may be either branched or unbranched. It should especially be noted that the catafusenes contain both the geometrically planar benzenoids and nonplanar helicenes.

Harary and Read [1] achieved a general mathematical solution for the numbers of catafusenes with given h values in terms of a generating function. Here, h is used to denote the number of hexagons. The "Harary–Read numbers", a term coined by Knop et al. [3], have been quoted many times [2–13]. They are also implied in a work of Herndon [14] and others [15]. Furthermore, the pertinent theory was exploited by Gutman [16].

It has become customary to classify polyhexes according to the symmetry groups to which they belong [13, 17]. For the numbers of *unbranched* catafusenes, a complete mathematical solution is known in terms of explicit formulas [18]. They give full information about the symmetry groups of interest, which are D_{2h}, C_{2h}, C_{2v} and C_s apart from D_{6h}, to which benzene ($h = 1$) belongs as the only (and trivial) catacondensed benzenoid. For the *branched* catafusenes, and hence also for the catafusenes in total, a corresponding symmetry distribution has not been worked out before; the possible symmetry groups for branched catafusenes are D_{3h}, C_{3h}, C_{2h}, C_{2v} and C_s.

The nonplanarity of helicenes is not taken into account when the symmetry group is defined. The system below, for instance, is attributed to the C_{2v} symmetry. The right-hand drawing is a dualist [18] representation.



In the present paper, a complete mathematical solution is given for the numbers of nonisomorphic catafusenes, including their distribution into the symmetry groups. This goal was reached by elementary combinatorial methods, without invoking generating functions, and without explicit use of Pólya's theorem, which Harary and Read [1] are referring to. Nevertheless, all the numbers for different classes of catafusenes from our main reference [1] were reproduced [19].

2. Symbols and definitions

Below we give a survey of applied symbols (in alphabetical order) with their definitions. Most of them are consistent with the notation of Harary and Read [1]. When there are differences, the corresponding Harary and Read symbols are given in brackets.

a	# hexagons in a single linear chain as a fragment of a catafusene;
A	# C_s catafusenes;
b	# hexagons in a fragment (arm) of a catafusene;
C	# C_{2h} catafusenes;
D	# D_{2h} catafusenes;
E	$F - G[F - h]$;
F	# hexagon-rooted catafusenes, where the reflection in a (mirror) plane is a forbidden symmetry operation;
G	# unrooted catafusenes, where reflection in a plane is forbidden [h];
h	# hexagons [n];
I	# D_{6h} catafusenes (trivial);

$M^{(a)}$ # catafusenes belonging to C_{2v}(a) [20], where the twofold symmetry axis (C_2) cuts edges;

$M^{(b)}$ # catafusenes belonging to C_{2v}(b) [20], where C_2 goes through vertices;

R # C_{3h} catafusenes;

T # D_{3h} catafusenes;

U # edge-rooted catafusenes;

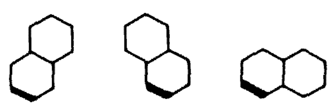
W # catafusenes with a plane of symmetry ($D_{6h} + D_{3h} + D_{2h} + C_{2v}$);

x: $x = h$ [n] for an edge-rooted catafusene, $x = h - 1$ [$n - 1$] for a hexagon-rooted catafusene;

ξ: variable in a generating function [x].

3. Catafusenes rooted at an edge

In an "edge-rooted" catafusene, a unique edge ("root-edge") in a unique hexagon is distinguished. It is not allowed to add any other hexagon adjacent or incident to the root edge. Neither is any symmetry operation allowed. Hence, for two hexagons for instance, there will be three rooted naphthalenes:



Here, the root edge is indicated in bold. Figure 1 shows the forms of the edge-rooted catafusenes (as dualists) for two and three hexagons. There are two types of these systems, S (single) and D (double) [1]. In fig. 1, all the systems but the last one are of the S type; the last one is D.

The numbers of edge-rooted catafusenes with x hexagons each, U_x, can be obtained for arbitrarily large x by the following recursive algorithm [1]:

$$U_0 = U_1 = 1, \quad U_2 = 3U_1,$$

$$U_{x+1} = 3U_x + \sum_{i=1}^{x-1} U_i U_{x-i}; \quad x = 2, 3, 4, 5, \ldots \quad (1)$$

The solution (1) is consistent with the generating function deduced by Harary and Read [1]. One has

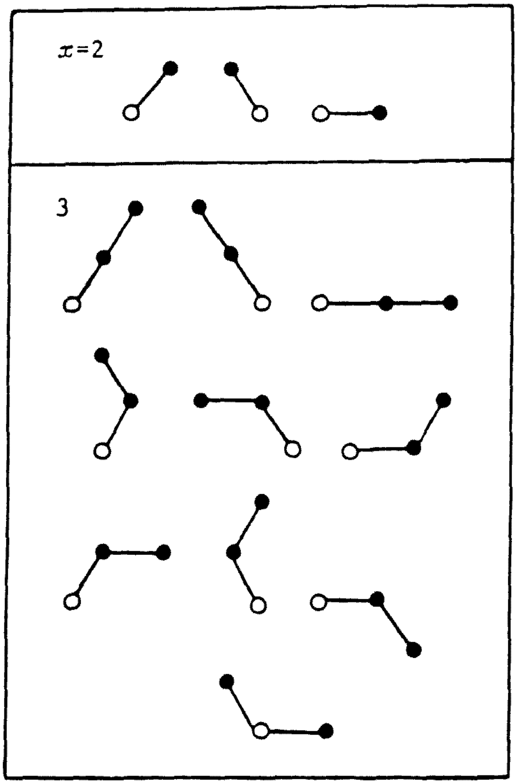


Fig. 1. Forms of the edge-rooted catafusenes with two and three hexagons. The dualist representation is employed. A white circle represents the hexagon containing the root edge.

$$1+(1/2)\xi^{-1}[1-3\xi-(1-\xi)^{1/2}(1-5\xi)^{1/2}]=\sum_{i=0}^{\infty}U_i\xi^i=1+\xi+3\xi^2$$
$$+10\xi^3+36\xi^4+137\xi^5+543\xi^6+2219\xi^7+9285\xi^8+39587\xi^9$$
$$+171369\xi^{10}+751236\xi^{11}+3328218\xi^{12}+14878455\xi^{13}$$
$$+670307855\xi^{14}+3040361705\xi^{15}+\dots\ . \tag{2}$$

The Harary and Read function was only modified by adding the constant term in order to take care of $U_0=1$. It is convenient to make this definition in view of the deductions in the following.

4. Unrooted symmetrical catafusenes

4.1. REGULAR HEXAGONAL SYMMETRY, D_{6h}

The trivial result reads:

$$I_1 = 1, \ I_h = 0; \qquad h > 1. \tag{3}$$

4.2. REGULAR TRIGONAL SYMMETRY, D_{3h}

The catafusenes of D_{3h} symmetry were treated by the same method as used in a computer algorithm [21] for enumeration of the catabenzenoids (without helicenes) of D_{3h} symmetry. The catafusene (like the catabenzenoid) consists of three identical arms attached to a central hexagon. Let the number of hexagons in one arm be

$$b = a + 2x, \tag{4}$$

where a is the number of hexagons in a single linear chain (acene). The total number of hexagons of the D_{3h} system is

$$h = 3b + 1. \tag{5}$$

All such non-isomorphic systems are obviously generated by taking all possible a values in conjuction with appropriate x values and in each case using the U_x edge-rooted catafusenes. Specifically, for a given b, use $a = b, \ b - 2, \ b - 4, \ldots$, down to 1 or 2, depending on whether b is odd or even, respectively. The corresponding x values (in reverse order) are $0, 1, 2, \ldots$ up to $(b - 1)/2$ or $(b - 2)/2$ for b odd or b even, respectively. In conclusion, the number of non-isomorphic D_{3h} catafusenes with h hexagons (T_h) becomes

$$T_{3b+1} = \sum_{i=0}^{\lfloor (b-1)/2 \rfloor} U_i; \quad b = 1, 2, 3, 4, \ldots \tag{6}$$

and finally in terms of h:

$$T_h = \sum_{i=0}^{\lfloor (h-4)/6 \rfloor} U_i; \quad h = 4, 7, 10, 13, \ldots,$$

$$T_h = 0 \qquad \text{otherwise.} \tag{7}$$

4.3. DIHEDRAL SYMMETRY D_{2h}

The treatment of the D_{2h} catafusenes is as simple as in the case of D_{3h} and similar to it. Let the number of hexagons be

$$h = a + 4x \tag{8}$$

when the system under consideration has a central linear chain of a hexagons. Then, $a = h,\ h-4,\ h-8,\ h-12, \ldots, a_{\min}$, where $a_{\min} = 2, 3, 4$ or 5. The corresponding maximum value of x appears as the upper summation index in the result given below for the number of catafusenes with D_{2h} symmetry, viz. D_h.

$$D_1 = 0,$$

$$D_h = \sum_{i=0}^{\lfloor (h-2)/4 \rfloor} U_i\,; \quad h = 2,3,4,5,\ldots\,. \tag{9}$$

Notice that the actual numbers D_h are the same as T_h, but distributed in a different way in relation to the h values.

4.4. CENTROSYMMETRY, C_{2h}

The case of C_{2h} symmetry is not treated here in detail for the sake of brevity, but was solved by the same kind of combinatorial reasonings as described above. The net result is:

$$C_1 = 0,$$

$$C_h = \tfrac{1}{2}\, U_{\lfloor h/2 \rfloor} - \tfrac{1}{2} \sum_{i=0}^{\lfloor (h-2)/4 \rfloor} U_i = \tfrac{1}{2}\left(U_{\lfloor h/2 \rfloor} - D_h\right); \quad h = 2,3,4,5,\ldots\,. \tag{10}$$

4.5. NON-REGULAR TRIGONAL SYMMETRY, C_{3h}

For the C_{3h} symmetry, the numbers (R_h) are the same as C_h but distributed in a different way in relation to h. It was found that:

$$R_h = C_{2(h-1)/3}; \qquad h = 4, 7, 10, 13, \ldots,$$

$$R_h = 0 \qquad \text{otherwise.} \tag{11}$$

4.6. MIRROR SYMMETRY OF THE TYPE C_{2v}(a)

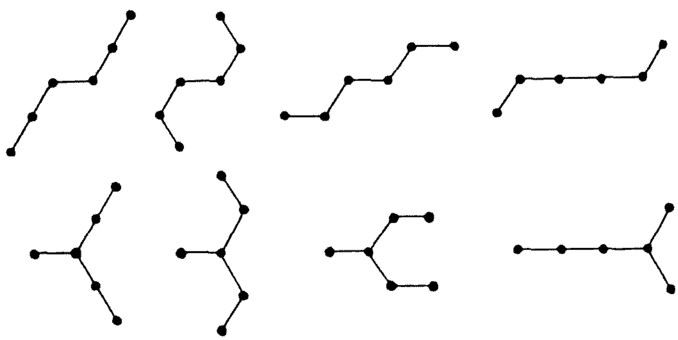
The catafusenes of C_{2v} symmetry are subdivided into two classes [20], identified by C_{2v}(a) and C_{2v}(b). In the former case (a), the twofold symmetry axis cuts edges (perpendicularly). Let the pertinent number of non-isomorphic catafusenes as a function of h be denoted by $M_h^{(a)}$. Below we write down the result which was obtained, and provide some explanations afterwards.

$$M_1^{(a)} = 0,$$

$$M_h^{(a)} = C_h - T_h, \qquad h = 2,4,6,8,\ldots, \tag{12}$$

$$M_{h+1}^{(a)} = C_h + U_{h/2} - T_{h+1}; \quad h = 2,4,6,8,\ldots .$$

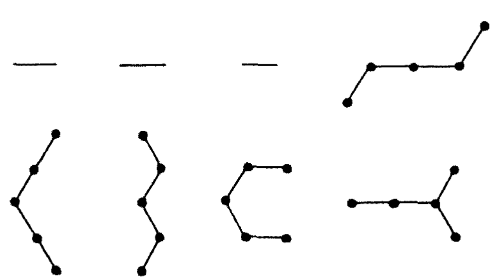
For even-numbered h, one finds the same number of catafusenes belonging to C_{2v}(a) and D_{3h} taken together on the one hand, and the number of C_{2h} catafusenes on the other. A one-to-one correspondence between the systems of the two classes can be established, as is exemplified for $h = 6$ below; $M_6^{(a)} = C_6 = 4$ ($T_6 = 0$). Dualists are employed:



C_{2h}

C_{2v}(a)

For odd-numbered h, one again finds a one-to-one correspondence between the C_{2v}(a) + D_{3h} and the C_{2h} systems. However, for $h > 1$ some C_{2v}(a) systems appear in addition, namely those which consist of two isomorphic edge-rooted catafusenes attached to one central hexagon. To take $h = 5$ as an example, one has $C_5 = 1$ and a corresponding C_{2v}(a) system. In addition come the $U_2 = 3$ systems built up from the edge-rooted naphthalenes (cf. fig. 1 and the accompanying text):



C_{2h}

C_{2v}(a)

4.7. MIRROR SYMMETRY OF THE TYPE C_{2v}(b)

The catafusenes belonging to C_{2v}(b) each have a twofold symmetry axis which passes through vertices. There is a perfect one-to-one correspondence between these systems and those of the C_{2h} symmetry; one only has to flip one of the fragments (an edge-rooted catafusene) around in a kind of a *cis/trans* isomerism. Therefore:

$$M_h^{(b)} = C_h. \tag{13}$$

4.8. TOTAL NUMBER OF CATAFUSENES WITH A PLANE OF SYMMETRY

$$W_h = I_h + T_h + D_h + M_h^{(a)} + M_h^{(b)}. \tag{14}$$

This quantity, which now is accessible through eqs. (3), (7), (9), (12) and (13), is consistent with the appropriate generating function from Harary and Read [1]:

$$\begin{aligned} \xi &= (1/2)\xi^{-2}(1+2\xi)[1-3\xi^2-(1-\xi^2)^{1/2}(1-5\xi^2)^{1/2}] \\ &= \sum_{i=1}^{\infty} W_i\,\xi^i = \xi+\xi^2+2\xi^3+3\xi^4+6\xi^5+10\xi^6+20\xi^7 \\ &\qquad + 36\xi^8+72\xi^9+137\xi^{10}+\ldots . \end{aligned} \tag{15}$$

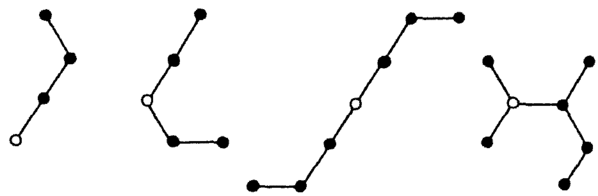
This expansion can easily be extended (up to W_{31}, actually) by mean of the coefficients given in eq. (2) and the relations [1]:

$$W_h = U_{h/2}, \quad W_{h+1} = 2W_h; \quad h = 2, 4, 6, 8, \ldots . \tag{16}$$

5. Catafusenes rooted at a hexagon

5.1. PARTITIONING

Harary and Read [1] have distinguished betwen four types of "hexagon-rooted" catafusenes: (i), (ii), (iii) and (iv). In such a system, a unique hexagon (the "root hexagon") is distinguished. The four types are exemplified below by means of the same systems as were depicted by Harary and Read [1], but here represented in terms of dualists. A white circle indicates the root hexagon.



The following formulas were found for the numbers within each category. For the sake of brevity, the results are given here without derivation, but at least those for types (i) and (ii) are obvious. The symbol x denotes the number of hexagons when the root hexagon itself is not counted.

5.2. TYPE (i)

$$F_{x+1}(\mathrm{i}) = U_x. \tag{17}$$

Here, the trivial value $U_0 = 1$ takes care of the root hexagon alone.

5.3. TYPE (ii)

$$F_1(\mathrm{ii}) = F_2(\mathrm{ii}) = 0,$$

$$F_{x+1}(\mathrm{ii}) = \sum_{i=1}^{x-1} U_i U_{x-i}; \quad x = 2,3,4,5,\ldots . \tag{18}$$

It should be understood that the reflection in a plane is a forbidden symmetry operation as a part of the definition of this class.

5.4. TYPE (iii)

$$F_1(\mathrm{iii}) = F_2(\mathrm{iii}) = 0,$$

$$F_3(\mathrm{iii}) = 1,$$

$$F_{x+1}(\mathrm{iii}) = \sum_{i=1}^{(x-1)/2} U_i U_{x-i}; \qquad x = 3,5,7,9,\ldots ,$$

$$F_{x+1}(\mathrm{iii}) = \tfrac{1}{2} U_{x/2}(U_{x/2}+1) + \sum_{i=1}^{(x/2)-1} U_i U_{x-i}; \quad x = 4,6,8,10,\ldots . \tag{19}$$

5.5. TYPE (iv)

$$F_1(\mathrm{iv}) = F_2(\mathrm{iv}) = F_3(\mathrm{iv}) = 0,$$

$$F_4(\mathrm{iv}) = 1,$$

$$F_{x+1}(\mathrm{iv}) = \tfrac{1}{3} U_{x/3}\left(U_{x/3}^2 + 2\right)$$
$$+ \sum_{i=1}^{(x/3)-1} \left[U_i^2 U_{x-2i} + U_i \sum_{j=i+1}^{x-2i-1} U_j U_{x-i-j} \right]; \quad x = 6, 9, 12, 15, \ldots; \qquad (20)$$

otherwise:

$$F_{x+1}(\mathrm{iv}) = \sum_{i=1}^{\lceil x/3 \rceil - 1} \left[U_i^2 U_{x-2i} + U_i \sum_{j=i+1}^{x-2i-1} U_j U_{x-i-j} \right].$$

5.6. TOTAL FOR HEXAGON-ROOTED CATAFUSENES

$$F_h = F_h(\mathrm{i}) + F_h(\mathrm{ii}) + F_h(\mathrm{iii}) + F_h(\mathrm{iv}). \qquad (21)$$

The individual numbers, obtained from eqs. (17)–(20), are listed in table 1 up to $h = 15$. The totals (F_h) are consistent with the appropriate generating function

Table 1

Partitioned numbers of hexagon-rooted catafusenes (mirror reflection not allowed)

h	F_h(i)	F_h(ii)	F_h(iii)	F_h(iv)
1	1	0	0	0
2	1	0	0	0
3	3	1	1	0
4	10	6	3	1
5	36	29	16	3
6	137	132	66	19
7	543	590	300	107
8	2219	2628	1314	543
9	9285	11732	5884	2709
10	39587	52608	26304	13246
11	171369	237129	118633	63918
12	751236	1074510	537255	306345
13	3328218	4893801	2447172	1461957
14	14878455	22395420	11197710	6959700
15	67030785	102943815	51473017	33092280

from Harary and Read [1]. It should not be necessary to reproduce the algebraic form of this function here; below we give only a part of its expansion.

$$\sum_{i=1}^{\infty} F_i \xi^i = \xi + \xi^2 + 5\xi^3 + 20\xi^4 + 84\xi^5 + 354\xi^6$$
$$+ 1540\xi^7 + 6704\xi^8 + 29610\xi^9 + 131745\xi^{10} + 591049\xi^{11}$$
$$+ 2669346\xi^{12} + 12131148\xi^{13} + 55431285\xi^{14} + 254539897\xi^{15} + \ldots \quad (22)$$

The individual numbers (table 1) may of course also be checked (as we have done) against appropriate parts of the generating function under consideration.

6. Unrooted asymmetrical catafusenes

6.1. AUXILIARY NUMBERS

Following Harary and Read [1], we have produced (in an alternative way) the numbers which bring us from F to G, where G pertains to all unrooted catafusenes where reflection in a plane is a forbidden symmetry operation. Then,

$$G_h = F_h - E_h, \quad (23)$$

where the E_h numbers are specified below, again without the derivation.

$$E_1 = E_2 = 0,$$
$$E_h = \sum_{i=1}^{(h-1)/2} U_i U_{h-i}; \qquad h = 3, 5, 7, 9, \ldots,$$
$$E_h = \tfrac{1}{2} U_{h/2}(U_{h/2} - 1) + \sum_{i=1}^{(h/2)-1} U_i U_{h-i}; \quad h = 4, 6, 8, 10, \ldots. \quad (24)$$

6.2. ASYMMETRICAL SYSTEMS C_s

$$A_h = \tfrac{1}{2}(G_h - W_h) - R_h - C_h. \quad (25)$$

7. Unrooted catafusenes in total

The final Harary–Read numbers are now, of course, obtained by:

$$H_h = \tfrac{1}{2}(G_h + W_h) = I_h + T_h + R_h + D_h + C_h + M_h^{(a)} + M_h^{(b)} + A_h. \quad (26)$$

Table 2

Numbers of non-isomorphic catafusenes, classified according to symmetry: I/D_{6h}, T/D_{3h}, R/C_{3h}, D/D_{2h}, C/C_{2h}, $M^{(a)}/C_{2v}$(a), $M^{(b)}/C_{2v}$(b), A/C_s, H/total

h	I	T	R	D	C	$M^{(a)}$	$M^{(b)}$	A	H
1	1	0	0	0	0	0	0	0	1
2	0	0	0	1	0	0	0	0	1
3	0	0	0	1	0	1	0	0	2
4	0	1	0	1	1	0	1	1	5
5	0	0	0	1	1	4	1	5	12
6	0	0	0	2	4	4	4	23	37
7	0	1	1	2	4	13	4	98	123
8	0	0	0	2	17	17	17	393	446
9	0	0	0	2	17	53	17	1600	1689
10	0	2	4	5	66	64	66	6486	6693
11	0	0	0	5	66	203	66	26694	27034
12	0	0	0	5	269	269	269	110818	111630
13	0	2	17	5	269	810	269	465890	467262
14	0	0	0	15	1102	1102	1102	1978032	1981353
15	0	0	0	15	1102	3321	1102	8481860	8487400

Table 2 shows the numbers of unrooted catafusenes, distributed over the different symmetry groups. It should be needless to say that the totals (H) match perfectly the results from the celebrated final generating function $H(\xi)$ of Harary and Read [1].

As the last equation, we give the summation formula for the total number of catafusenes in closed form (for H_h when $h \le 5$, see table 2). It is the counterpart of the $H(\xi)$ generating function in the sense that

$$H(\xi) = \sum_{i=1}^{\infty} H_i \xi^i, \tag{27}$$

$$\begin{aligned} H_h = {} & \tfrac{1}{2} U_{h-1} + \tfrac{1}{8}[1-(1)^h] U_{\lfloor (h-1)/2 \rfloor}(3U_{\lfloor (h-1)/2 \rfloor} + 1) \\ & - \tfrac{1}{8}[1-(-1)^h] U_{\lfloor h/2 \rfloor}(U_{\lfloor h/2 \rfloor} - 1) + \tfrac{1}{4}[3-(-1)^h] U_{\lfloor h/2 \rfloor} \\ & + \tfrac{3}{2} \sum_{i=1}^{\lfloor (h/2)-1 \rfloor} U_i U_{h-i-1} - \tfrac{1}{2} \sum_{i=1}^{\lceil (h/2)-1 \rceil} U_i U_{h-i} \\ & + \tfrac{1}{6}(\lfloor (h-1)/3 \rfloor - \lfloor (h-2)/3 \rfloor) U_{\lfloor (h-1)/3 \rfloor}(U^2_{\lfloor (h-1)/3 \rfloor} + 2) \\ & + \tfrac{1}{2} \sum_{i=1}^{\lfloor (h-2)/3 \rfloor} U_i \left[U_i U_{h-2i-1} + \sum_{j=i+1}^{h-2i-2} U_j U_{h-i-j-1} \right]; \quad h > 5, \end{aligned} \tag{28}$$

where the U_x numbers are given by eq. (1).

8. Conclusion

In the present work, the "Harary–Read numbers" for catafusenes are re-derived for the first time in an alternative way, twenty years after the pioneering work of Harary and Read [1]. As an original contribution, the numbers are divided according to the symmetries of the systems in question. The task was accomplished by elementary combinatorial methods, which led to summation formulas. It is not claimed that the present methods are "better" or even simpler than the applications of generating functions, which were exploited by Harary and Read [1]. It is supposed, however, that the present methods provide a useful alternative.

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[19] There is obviously a minor misprint in the table of Harary and Read [1]: G_{10} (our notation; see section 2) should be 13249.
[20] I. Gutman and S.J. Cyvin, *Introduction to the Theory of Benzenoid Hydrocarbons* (Springer, Berlin, 1989).
[21] T. Tošić, Z. Budiman, J. Brunvoll and S.J. Cyvin, J. Mol. Struct. (THEOCHEM) 209(1990)289.