

Rhombellanic Crystals and Quasicrystals

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ABSTRACT

Design of some crystal and quasicrystal networks, based on rhombellane tiling, is presented. [1,1,1]Propellane, is a synthesized organic molecule; its hydrogenated form, the bicyclo[1.1.1]pentane, may be represented by the complete bipartite graph $K_{2,3}$ which is the smallest rhombellane. Topology of translational and radial structures involving rhombellanes is described in terms of vertex symbol, connectivity sequence, ring sequence and map operations relating structures to their seeds. It is shown, by alternating sum of ranked substructures, that radial structures represent complex constructions of higher rank. Basic properties of rhombellanes, coloring included, are outlined.

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1 INTRODUCTION

[1,1,1]Propellane is an organic molecule, first synthesized by Wiberg and Walker in 1982 [1]. By IUPAC rules, it is named Tricyclo[1.1.1.0^{1,3}]pentane, a hydrocarbon with formula C_5H_6 and three rings of three atoms. The hydrogenated form of propellane, C_5H_8 , eventually named bicyclo[1.1.1]pentane, has only rhomb/square rings; it can be represented by $K_{2,3}$ - the complete bipartite graph, which is the smallest rhombellane. The two bridge carbon atoms can be functionalized, e.g., by bromine or COOH, or even by repeating the $K_{2,3}$ motif, as in the polymer called staffane [2].

Rhombic polyhedra are known as aesthetic appeal objects, of mathematical interest [3]; the well-known triacontahedron, the dual of Archimedean

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icosidodecahedron, has 30 rhombic faces. In the book “Multi-shell polyhedral clusters” [4], the cluster C_{152} was described consisting of $K_{2,3}$ units, which are not polyhedra cf. Steinitz Theorem [5] but tiles [6].

Design of rhombellanes is made by a general procedure [7], achieved as follows: join by a point (called “rbl-point”) the two vertices lying opposite diagonal in each rhomb of an all rhomb-map (i.e. the zero-generation, Rh_0). Then, add new vertices opposite to the parent vertices and join each of them with the rbl-vertices lying in the proximity of each parent vertex, thus local Rh-cells being formed. The process can continue, taking the envelope Rh_n as “ Rh_0 ” for Rh_{n+1} , in this way shell by shell being added to the precedent structure. Since the two diagonals may be topologically different, each generation may consist of two isomers.

The paper is organized as follows: after an introduction, construction of some periodic rhombellane-consisting structures is presented; in the third section, non-periodic radial structures are discussed; the fourth section details the rhombellanic character, in mathematical chemistry terms; in the fifth section, a graph coloring problem related to rhombellanes is exposed; conclusions and references will close the paper.

2. PERIODIC RHOMBELLANIC STRUCTURES

According to Steinhardt definition [8], crystals are highly ordered structures, with atomic clusters repeated periodically, in three independent directions of the space, and showing an essentially discrete diffraction diagram; the symmetry of infinite crystal lattices is completely described by the 230 symmetry groups of the space.

Starting from the simplest crystal network, namely the simple cubic *pcu* net, of which repeating unit is a cube C , it is possible to build a variety of triple periodic structures.

Let first locate a point/atom in the center of cube and join it with all the corners of cube; the obtained unit is referred here as $CP^{8,9}$ (Figure 1, left), P^8 meaning a point of connectivity 8. By translating this unit along the three coordinate axes results in a “body centered cubic” *bcc* network, including both *pcu* and *bcu* networks; by this reason, it is named here *pcu-bcu* (Figure 2, left). Second, cut-off, in an alternating manner, four of the edges emerging from the central point to the corners of cube; the unit thus obtained is named $CP^{4,9}$ (Figure 1, middle) while the network resulted from it by a simple translation is denoted *pcu-dia* (Figure 2, middle). Third, translate CP^4 unit along the three coordinate axes, each step rotated 90° , thus resulting the network called here *pcu-flu* (Figure 2, right); its repeating unit consists of eight CP^4 units, with a total of 35 points/atoms.

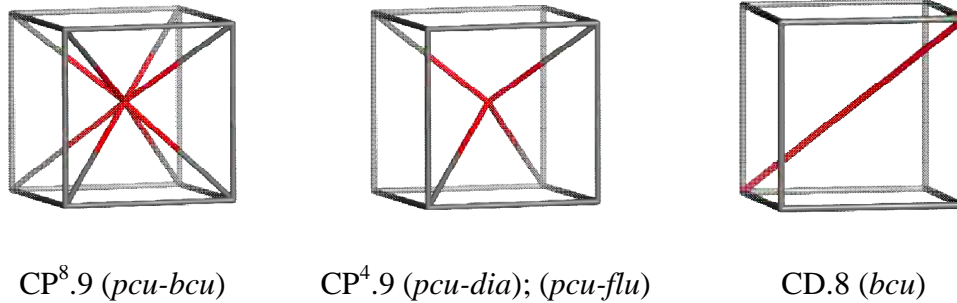


Figure 1. Seeds for three periodic networks.

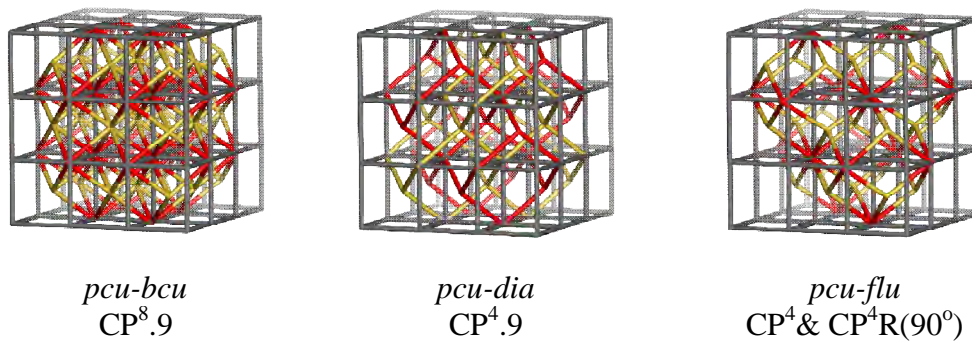


Figure 2. Networks superposed over the simple cubic net *pcu*; seeds are indicated on the bottom row.

The *bcu*, *dia* and *flu* nets alone (see [9] for symbols), resulted by deleting the *pcu* net (Figure 2), are illustrated in Figure 3. Also, *bcu* can be generated by translating the unit CD.8 (Figure 1, right), a diagonalized cube, representing a substructure of the unit $Rh_{12}P^8.15$ (Figure 4, top, middle).

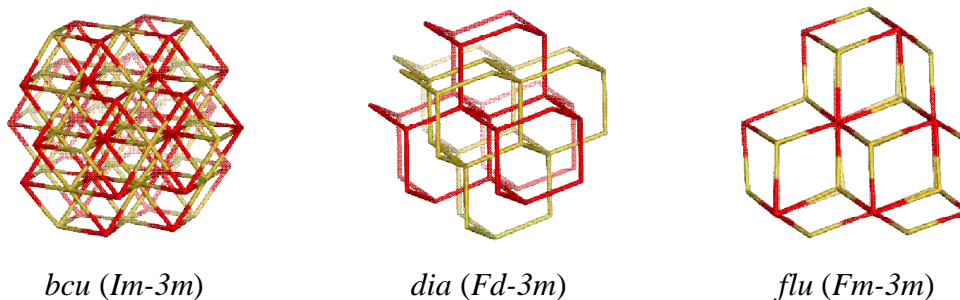


Figure 3. Periodic nets envisaged by deleting the simple cubic net *pcu* in Figure 2.

The void of *flu* net is the rhombic dodecahedron $Rh_{12}.14$ (Figure 4, top, left), a space filler. By doping Rh_{12} with a body centered atom, P^8 , results in $Rh_{12}P^8.15$, a cluster of rank $k = 5$ (see Table 1S, Supplementary material); it is the seed of *bcu* net (Figure 5, top, middle and right); relation between the two networks is illustrated in Figure 4. Any atom of *bcu* is retrieved in the *pcu* (i.e., twin/entangled *pcu*) net (and the reciprocal is true); it is clear that Figure 2, left, shows the *bcu* net with the rectangular edges of *pcu* also represented. However, any of *flu* atoms belongs to the (twin) *pcu* but the reciprocal is not true.

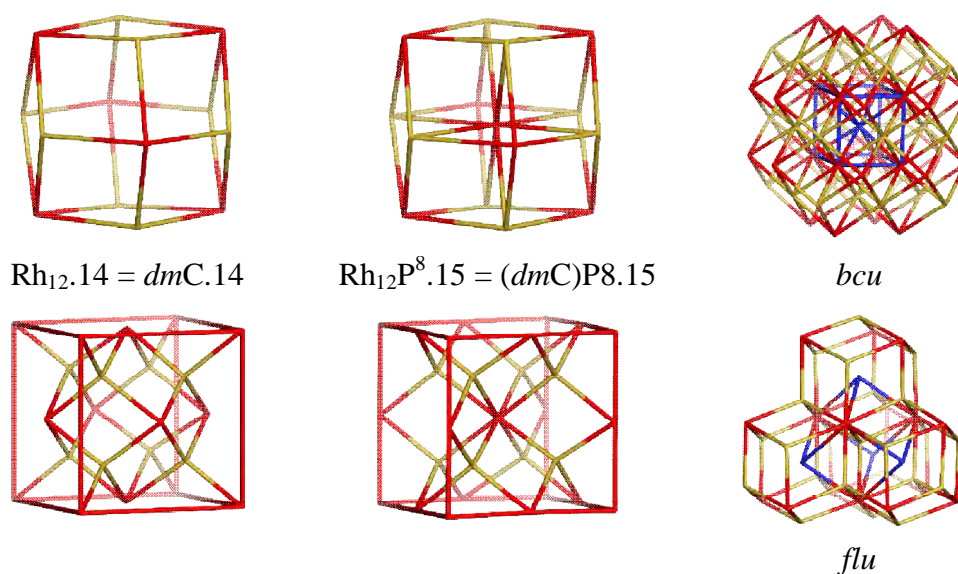


Figure 4. Doping by a point/atom the seed of *flu*, Rh_{12} (top, left), becomes $Rh_{12}P^8.15$ (top, middle), the seed of *bcu* (top right); the void Rh_{12} and its complement (within the *pcu* frame) form the *flu* net (bottom).

Similarly, the net *pcu-dia* (Figure 2, middle), of which seed is $CP^4.9$, is in fact the twin *dia* net, of $Fm-3m$ space group: a “face centered cubic” *fcc* net is entangled with its self-dual net; the two nets are displaced along the body diagonal of the cube by one quarter of the diagonal length, as illustrated in Figure 5. Any atom of (twin) *dia* is retrieved in the (twin) *pcu* net (and the reciprocal is true); in the retrieved *pcu* net, the atoms of the two *dia* nets alternate in populating the cubic net, as in the cube bipartite coloring (Figure 6). If rotates 90° to each other (and identifies the superposed points) *dia-dia* changes to the *flu* net (Figures 3 and 4).

Triple periodic networks can be characterized by sequences of vertex connectivity, as given in the crystallographic databases [9]. Sequences of a given topological property are counted as rows in layer/shell matrices, LM/ShM [10,11]; in this case, LC is the layer of connectivity matrix, which is taken up to the distance ten from the chosen vertex. In addition, if the strong rings surrounding vertices are considered, the layer of rings matrix LR [12] can be obtained; the characterization of a triple periodic network is (for the first time) more complete. Lists of such data for the nets: *pcu*, *bcc*, *fcc*, *dia*, *flu*, *pcu-bcc*, *pcu-dia* and *pcu-flu* are given elsewhere (Tables 2S and 3S – Supplementary material). (The figure count for the seeds of the discussed networks is given in Table 1S– Supplementary material).

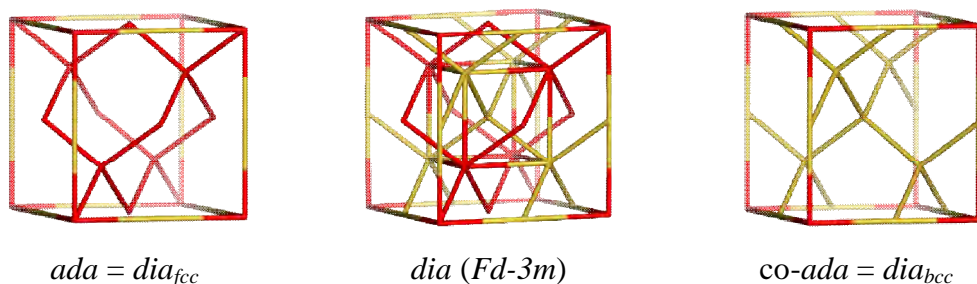


Figure 5. Diamond net substructures entangled within the *pcu* frame: $dia = \cup(fcc; bcc)$.

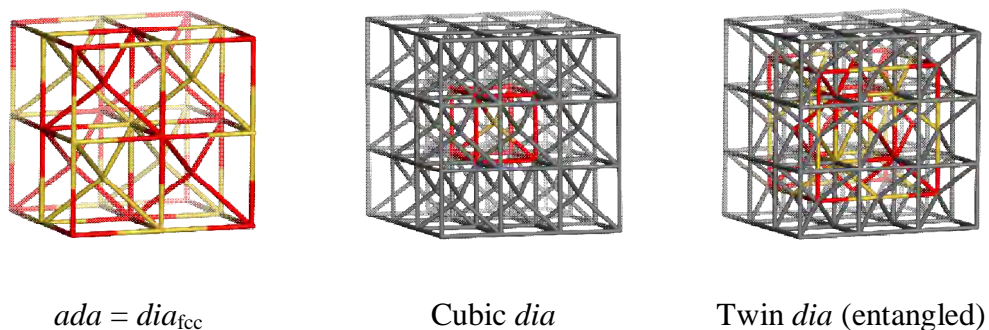


Figure 6. Two *dia*-nets complementarily occupy the same space generated by the CP^4 seed: there is only one *ada* unit (red) and one *co-ada* (yellow – left); in the cubic *dia* net, the space filler is only CP^4 unit (red – middle); in the retrieved *pcu* net, the atoms of the two entangled *dia* nets alternatively populate the cubic net, as in the bipartite coloring (right).

3. RHOMBELLANIC RADIAL STRUCTURES

Quasicrystals are finite aperiodic structures, with long-range positional and orientational order [8]. Among the rotational symmetries, 2-, 3-, 4- and 6-fold axes are allowed in crystals, while 5-, 7- and all higher (non-crystallographic) rotational symmetries are encountered in quasicrystals. Atomic clusters are repeated in a complex, non-periodic pattern; electron diffraction shows sharp patterns, as found experimentally by Shehtman [13] the Nobel prize winner in 2011. Radial structures with rhombellanic characteristics can be obtained by applying iteratively the “rhombellation” procedure, described in the introductory section. The procedure is illustrated in Figure 7, starting from the cube.

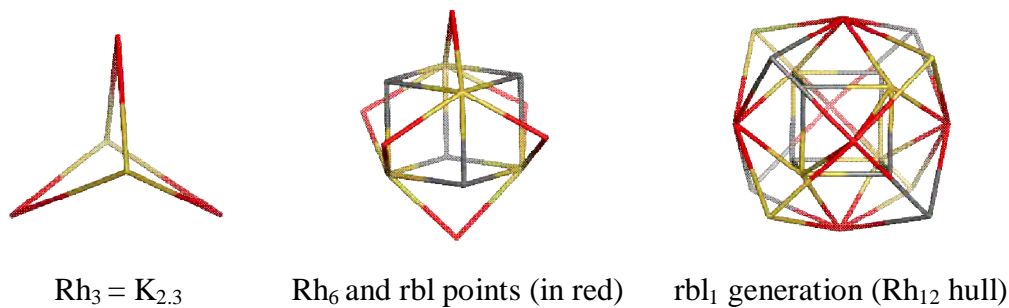


Figure 7. Rhombellane Rh₃ and rhombellation starting from the cube (*i.e.*, Rh₆).

The new envelope Rh_{*n*+1} has twice the number of rhombs in the precedent Rh_{*n*} envelope; the number of vertices in a new generation is counted iteratively as: $v_{n+1} = v_n + 2h_n + 2$, with $v = |V(G)|$ being the number of vertices, h_n the number of rhombs in the hull of *n*-generation (embedded in the sphere) and 2 is the Euler characteristic (see below) of the sphere. Referring to the zero-generation, Rh₀, the actual number of vertices can be obtained by the formula:

$$v_n = 2(n + 1) + h_0(2^{n+1} - 1).$$

Radial series can be characterized, as the crystal structures, by shells of connectivity LM and shells of rings around vertex LR matrices (see Supplementary material, Table 4S).

About space dimensionality or ranking, as defined by Schulte [14], each rhombellane generation (*i.e.* shell) can be seen as a cluster of rank $k = 4$ (Table 1); then, two such shells share a common 3-facet, which is a sphere tessellated by rhombs f_4 , a true cell (Figure 8). It means, a shell pair (1;2) or (2;3) are structures of rank $k = 5$. Further, a pair $\{(1;2);(2;3)\}$ will share a shell of rank $k = 4$ (in this

case, the shell (2)); thus, the structure 3_{full} , (Table 1, bottom), bonded by two facets of rank $k = 5$, is a structure of rank $k = 6$ and the process can continue.

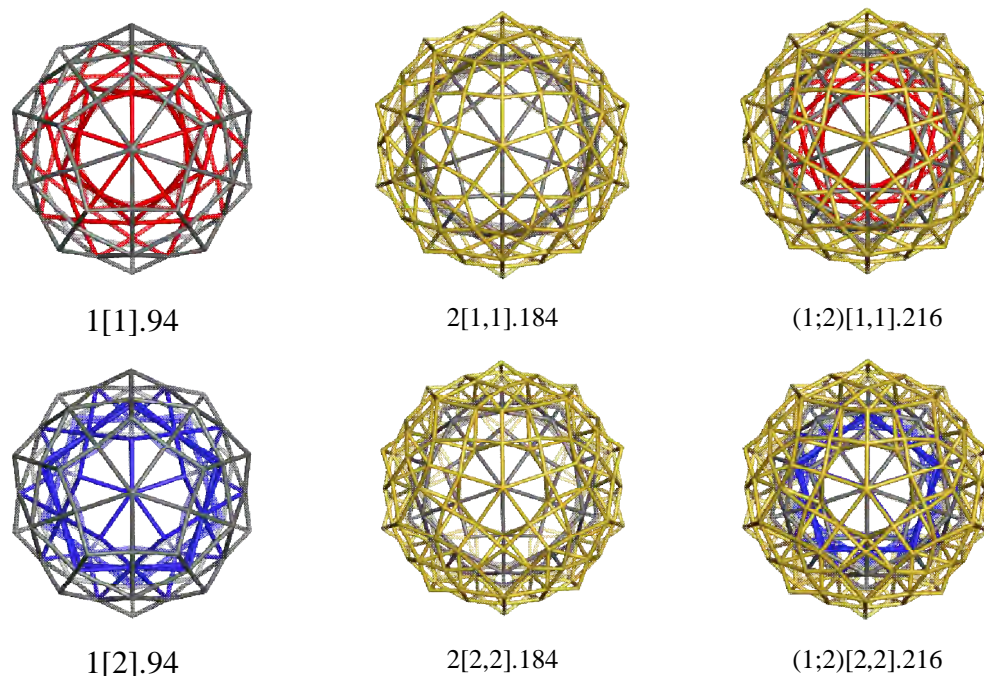


Figure 8. Facets of the shell pair (1;2) of rhombellanes $rb_l_n(\text{Rh}_{30})$.

Table 1. Figure count for $\text{Rh}_{30}rb_l_n[1, \dots, 1]$; $K_{2,3}^* = f_4/3$; M = No. (inner + outer) cells.

$[1, \dots, 1]$	v	e	f_4	$K_{2,3}$	$K_{2,4}$	$K_{2,5}$	Rh_8	Rh_{10}	$K_{2,3}^*$	M	3	4	5	6
1	94	240	270	20	0	0	0	12	90	2	124	0	(30;60)	
2	184	480	540	20	0	12	30	0	180	2	244	0	(60;120)	
3	364	960	1080	20	30	12	60	0	360	2	484	0	(120;240)	
(1;2)	216	600	750	40	0	12	30	12	270	2	366	2	2	-
(2;3)	426	1200	1470	40	30	24	60	0	540	2	696	2	2	-
3_{full}	458	1320	1710	60	30	24	90	12	630	2	848	2	2	0

Note, in Table 1, the presence of $K_{2,n}$ complete bipartite graphs and related rhombic cells Rh_n ; also note the count of f_4 (pair $(\text{Rh}_n, \text{Rh}_{n+1})$ at the top of #5 and #6 columns). For the series $[2, \dots, 2]$, see Table 5S, (Supplementary material). Euler characteristic χ [15] of a surface S can be calculated as an alternating sum of figures, of rank k : $\chi(S) = f_0 - f_1 + f_2 - f_3 + \dots$.

4. RHOMBELLANIC CHARACTER

Proposition [7]. A structure is a rhombellane if all the following conditions are obeyed: (a) All strong rings are squares/rhombs; (b) Vertex classes consist of all non-connected vertices; (c) Omega polynomial has a single term: $1X^{|E|}$; (d) Line graph of the original graph shows a Hamiltonian circuit; (e) Structure contains at least one $K_{2,3}$ subgraph.

Cube (actually Rh_6) is an all-square graph and Hamiltonian; its line-graph, the cuboctahedron, is also Hamiltonian but its Omega polynomial [16,17]: $\Omega(C) = 3X^4$, meaning not all of its edges are topologically parallel; also, the vertices of cube form a single vertex class and thus cannot be disconnected.

Triacanthedron, Rh_{30} , has all-square rings, all non-connected vertex classes but not $1X^e$ Omega polynomial unique term and no Hamiltonian circuit of its lines. Rhomb Icosahedron, Rh_{20} , has not all classes of non-connected vertices.

Omega polynomial is defined as: $\Omega(x) = \sum_k m x^s$, m being the number of opposite edge strips, ops , of length s , in a graph G . There are graphs with a single ops , which is a Hamiltonian circuit. For such graphs, Omega polynomial has a single term: $\Omega(x) = 1x^s$; $s = e = |E(G)|$, in other words, "all the edges in G are topologically parallel". However, Hamiltonicity is an *NP* complete problem, being taken here as a corollary of a single ops in Omega polynomial; however, not all the graphs having a Hamiltonian circuit have all the edges topologically parallel (see the case of cube and cuboctahedron).

The smallest rhombellane Rh_3 is $K_{2,3}$, the complete bipartite graph (corresponding to the molecular graph of C_5H_8 , bicyclo[1.1.1]pentane); all $K_{2,n}$ graphs fulfill all the above conditions. Any $K_{2,n}$ graph consists of $n(n-1)(n-2)/6$ $K_{2,3}$ substructures. There are rhomb-tessellated cages that fulfil the first four criteria but do not contain any $K_{2,3}$ substructure.

Further, there are graphs with more than two vertex classes obeying the above conditions. Rhombellation operation provides such graphs, with n shells/generations, when applied iteratively. The rbl-vertices added in the first step of any new generation are disjoint with respect to each other while in the second step they are joined by means of new vertices superposed on the parent vertices (thus not connected, neither to the parent vertices nor to themselves); this construction provides classes of vertices non-connected to each other within a same class. Rhombellanes represent n -partite graphs, both by topology and coloring (see below). Rhombellanic crystal networks also fulfill all the five above criteria: among the discussed network, only the *dia* net (as the superposed *pcu-dia*) is full rhombellanic, whereas *pcu-bcu* has triangles while *flu* does not cover all

points/atoms in *pcu*. Accordingly, only the CP⁴.9 seed show a full rhombellanic character.

Corollary. *In a finite molecular rhombellane, with vertex classes consisting of distinct atom types, there are only polar bonds while covalent non-polar bonds may not exist.*

5. COLORING PROBLEM

The chromatic number *Ch* of a graph is the smallest number of colors needed to color its vertices so that no edge has the both endpoints colored the same [18]. Several graph constructions have been proposed about graph coloring [19–22]; two of them are more related to our proposed construction:

Mycielski’s Theorem ([23], 1955). For any integer $n > 1$, there exists a triangle-free n -chromatic graph.

Zykov’s Theorem ([24], 1949): There exist triangle-free graphs with arbitrary large chromatic number.

Hamiltonicity and other properties of triangle-free graphs transformed by Mycielski’s construction were discussed in [25,26]. Note that, the 4-polytope 24-Cell is three-colored, its medial (i.e., line-graph) C₉₆ is four-colored, its face-dual is also four-colored; however, these graphs have a single topological vertex class; it means, the coloring does not superposes over topology. Also, bipartite graphs (i.e. graphs with all even size cycles) have $Ch = 2$ but may have more than two topological vertex classes. In rhombellanes, topology superposes over coloring; for $rb1_1(C).22$, we found $Ch = 5$; for $rb1_2(C).48$, $Ch = 8$.

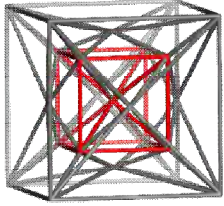
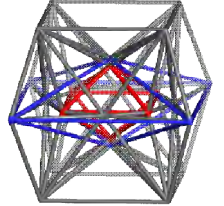
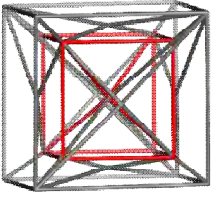
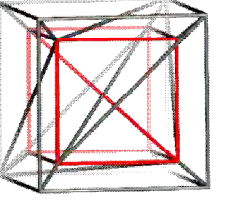
			
$Q_4.8CP^8.24 = 24\text{-Cell}$ $Ch=3, Cls=1$	$Q_4.8CP^8.24$ Torus $T_{4,4}$ embedding	$Q_4.8CP^4sa.24$ $Ch=2, Cls=2$	$Q_4.CD.16$ $Ch=16, Cls=16$

Figure 9. Hypercube Q_4 derivatives.

Jensen and Royle [27] provided an easy construction of a 22-vertex graph (from the Grötzsch graph) and an easy proof that the result is triangle-free and 5-chromatic. By repeating two times Mycielski's procedure, a 45-vertex, triangle-free, 6-chromatic graph was obtained; it is unknown if a smaller such graph exists [18] (however, graphs of 44 or 43 vertices were questioned [28]). In this light, our results are correct, with respect to chromatic number and our procedure seems to be simpler than those already published.

Rhombellation operation provides triangle-free graphs with arbitrarily large chromatic number. Figure 9 illustrates three derivatives of the hypercube Q_4 – Tesseract, in four representations: (i) $Q_4.8CP^8.24=24$ -Cell ($Ch=3$; $Cls=1$); the construction is made in the idea of cube-derivatives CP^8 , CP^4 and CD , used as network seeds (Figure 1) and seems to be a new way to build the 24-Cell 4-polytope [7]; it has $Ch = 3$ but is not rbl in character (it contains triangles and has a single class of vertices, thus cannot be disconnected); (ii) The object $Q_4.8CP^8.24$ embedded in the torus $T_{4,4}$; (iii) $Q_4.8CP^4.sa.24$, a syn-anti isomer with $Ch = 2$ and $Cls=2$; it has a rbl character and (iv) $Q_4.CD.16$, a diagonalized hypercube having $Ch=V(G)=16$ (i.e., the number of atoms/vertices in the molecule/graph); in other words, each class consists of singular vertices, clearly disconnected, as they belong each to different classes; this is also a rhombellanic structure, obeying all the five rbl criteria. Topology of these Q_4 -derivatives is given in Tables 6S to 8S (Supplementary material).

Vertex classes were computed by our Nano-Studio software [29], as centrality indices, and confirmed by permutations in the adjacency matrix of graphs, performed by Mathematica [30].

6. CONCLUSIONS

Rhombellane, Rh_3 or $K_{2,3}$, is the smallest tile with rhombic rings/faces; it represents a real chemical molecule. Generalized rhombellanes, designed by the rhombellation procedure, have non-connected vertex classes (of interest in graph coloring); all the edges are topologically parallel (as shown by the single term Omega polynomial, further involving Hamiltonian circuits visiting their edges) and contain at least one $K_{2,3}$ subgraph.

For some well-known triple periodic crystal networks, like *pcu*, *bcu* or *dia*, rhombellanes enable a deeper description, helpful in understanding relations among networks apparently not related. Cube-like molecules or crystal networks have been reported [31,32]. For the first time in literature, crystals and quasicrystals were characterized by sequences of strong rings around atoms.

Exploring network seeds led to a new building way of the 4-polytope 24-cell. Radial structures, generated by propellation are ordered (yet hypothetical) structures of higher rank.

Rhombellanes represent a new class of structures, with promising properties, both in theory and applications.

Supplementary Material. Available on request, at www.esmc.ro.

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کریستال‌ها و شبه‌کریستال‌های (مبلائی)

ادیتور (ابطا : علیرضا اشرفی)

چکیده

طرح برخی از شبکه‌های کریستال و شبه‌کریستال، بر پایه کاشی‌کاری رمبلائی، ارائه شده است. پراپلائی [1,1,1] یک مولکول آلی ترکیبی است؛ فرم هیدروژنه آن، پانتان [1,1,1] دوچرخه‌ای، احتمالاً توسط گراف کامل دو قسمتی $K_{2,3}$ که کوچکترین رمبلائی است، نشان داده می‌شود. توپولوژی ساختارهای انتقالی و شعاعی شامل رمبلائی‌ها برحسب نماد رأس، توالی اتصال، توالی حلقه و عملیات طرح مرتبط با بذرهایشان، توصیف می‌شود. به‌وسیله مجموع تناوبی زیرساختارهای رتبه‌بندی شده نشان داده می‌شود که ساختارهای شعاعی، ترکیب‌های مختلط رتبه بالاتر را نشان می‌دهند. ویژگی‌های اساسی رمبلائی‌ها، شامل رنگ آمیزی، مشخص می‌شوند.

لغات کلیدی: رمبلائی، کریستال، شبه‌کریستال، توپولوژی، ساختار رتبه بالاتر