

Bead Sculptures and Bead-Chain Interlocking Puzzles Inspired by Molecules and Nanoscale Structures

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Abstract

Mathematical beading can be utilized for the construction of aesthetically pleasing sculptures inspired by a rich variety of shapes and forms in the nanoworld. Depending on the shapes of beads utilized, the resulting bead sculptures can be classified into two broad categories: (1) hard-sphere open packing models based on spherical beads, which include finite fullerenes, carbon nanotubes, carbon nanotori, curved three-dimensional graphitic surfaces, and space-filling tetrahedral zeolite structures and many inorganic coordination complexes such as extended metal atom chains; (2) truss-like frameworks constructed from tubular bugle beads, which can mimic the polyhedral representations of microscopic inorganic structures and many macroscopic space frames. Moreover, the hard-sphere open packing model for any polyhedral hydrocarbon of general formula $C_{2n}H_{2n}$ can be achieved by cross-linkings of n pre-made five-bead chains without using any beading process. The assembly processes of certain symmetric polyhedral molecules, such as three Platonic hydrocarbons including tetrahedrane, cubane, and dodecahedrane, are similar to solving interlocking puzzles. Hence, these bead-chain models can also be regarded as a new type of take-apart put-together puzzle.

Introduction

Chemistry is the scientific discipline that deals with compositions, structures, properties of substances and the changes they undergo. Three dimensional structures depicting the arrangements of atoms inside a molecule or the larger-scale hierarchical organization of constituents of nanomaterials are particularly important for a thorough understanding of their physical and chemical properties. Through the advancement of various experimental techniques such as X-ray diffraction, electron microscopy, a large variety of structures and organizations of molecules, aggregates and nanomaterials have been determined. Model building, which has been proven to be a powerful approach for unravelling many important molecular structures in the past, also provides a simple way for understanding spatial relationships for both novices and experts alike.

In this paper, we describe two general approaches to the construction of bead models of molecules and nano structures. [1, 2, 3] In the first approach, we apply systematically the standard right angle weave (RAW) technique (a.k.a. figure-eight stitch) commonly used in the beading community for the construction of a large variety of sculptures inspired by nanoscale molecular structures. We have successfully built many interesting bead sculptures including the 3-connected 3D nets such as cage-like fullerenes, carbon nanotori, helically coiled carbon nanotubes, carbon torus knots, high-genus fullerenes, hyperbolic fullerenes, and triply periodic minimal surfaces, together with many 4-connected 3D nets such as diamond, ices, zeolites, and silicate minerals. [3, 4]

These bead models based on spherical beads can be regarded as the macroscopic realization of valence sphere models for the corresponding molecules, in which the spatial arrangement of spherical beads approximates the three-dimensional electron density distributions. The hard-sphere repulsion among beads and elastic cords that hold beads together mimics the nature of chemical bonds in alkanes, fullerenes, zeolites and many other nanostructures. The saturation and directional character of chemical affinity fall out naturally in bead models. Moreover, constructing the bead model of a molecule with mathematical beading can

be regarded as an analog computation. The result of the computation is the molecule's approximate three-dimensional electron density profile. [5, 6] In addition to spherical beads, we have also utilized tubular bugle beads for constructing many skeletal polyhedral models that can be regarded as physical models of microscopic inorganic structures and macroscopic octet-truss systems. The nodes in these skeletal structures can have connectivity or coordination numbers up to twelve, which is common in many octet-truss structures. [7, 8]

The second approach for the construction of bead models involves no beading at all. Constructing a bead sculpture with mathematical beading technique is usually a slow and tedious process. One needs not only to become familiar with the basic beading techniques, but also to know the algorithmic details for a specific sculpture. While the eventual completion of an aesthetically pleasing sculpture may prove that the effort of the whole process is worthwhile, many repetitive beading steps require a great deal of patience and practice. Therefore, it would be interesting if one can avoid the beading processes, but still keep the essential idea of the hard-sphere open packing in the bead models. Indeed, it is possible to simplify the complicated task of beading for certain molecules into two stages: First, the preparation of the simplest possible beaded structures, namely one-dimensional chains that consist of n beads; second, the assembly of these pre-made bead chains through suitable cross-linkings. The resulting cross-linked bead-chain models, if correctly manipulated, can be utilized to represent valence sphere models of various linear, ring-like, tree-like, and polycyclic alkane molecules, together with many inorganic systems such as perovskite, and so on.

A particularly interesting family of molecules that can be constructed by bead chains is the polyhedrane compound, a cage-like hydrocarbon molecule of general formula $C_{2n}H_{2n}$, which has a skeleton corresponding to a polyhedron. A few examples are tetrahedrane, cubane, dodecahedrane, and related prismanes. Moreover, the construction processes for these symmetric compounds have a strong resemblance to solving nontrivial interlocking puzzles such as Burr puzzles: namely starting from a given set of bead chains, each containing a specific number of beads. The goal is to assemble them into a target molecular structure through a sequence of cross-linkings. For instance, two bead chains, each consisting of five beads, can be assembled into the valence sphere model of a tetrahedrane molecule, while four five-bead chains can be put together into the valence sphere model of a cubane molecule. We call this new kind of puzzles as Bead-Chain Puzzles (abbreviated as BC-Puzzles). [9]

Techniques and Algorithms: Degree Three Polyhedra

In this section, we describe the basic beading techniques and procedures for making a bead model. [5, 10] First of all, one needs to prepare the raw materials needed for constructing beaded molecules which are just beads and threads (fishing lines or elastic cords). Beads are made in many materials and come in different colors, textures, and sizes ranging from several millimeters to about a centimeter and usually contain a thin channel for stringing. The sizes of beads we used for constructing bead sculptures in this paper are mostly around 6 mm to 10 mm. Every bead sculpture shown here is constructed by beads with the same size. In principle, the same beading technique can be easily generalized to make sculptures consisting of different sizes of beads. In order to construct a robust bead sculpture, it is crucial to choose threads with right thickness. A simple criteria for a degree three (cubic) polyhedral graph such as a fullerene is to choose the thickness of threads in such a way that they can go through the thin channel of beads at least three times, but no more than four times. The reason for this choice will become clear after we describe the techniques for making a bead sculpture.

Figure 1 gives the complete instruction on using the RAW technique to make a beaded cube which consists of twelve beads sitting on the midpoints of twelve edges. The whole procedure has six steps and each step consists of creating a four-member ring that represents a square face of a cube.

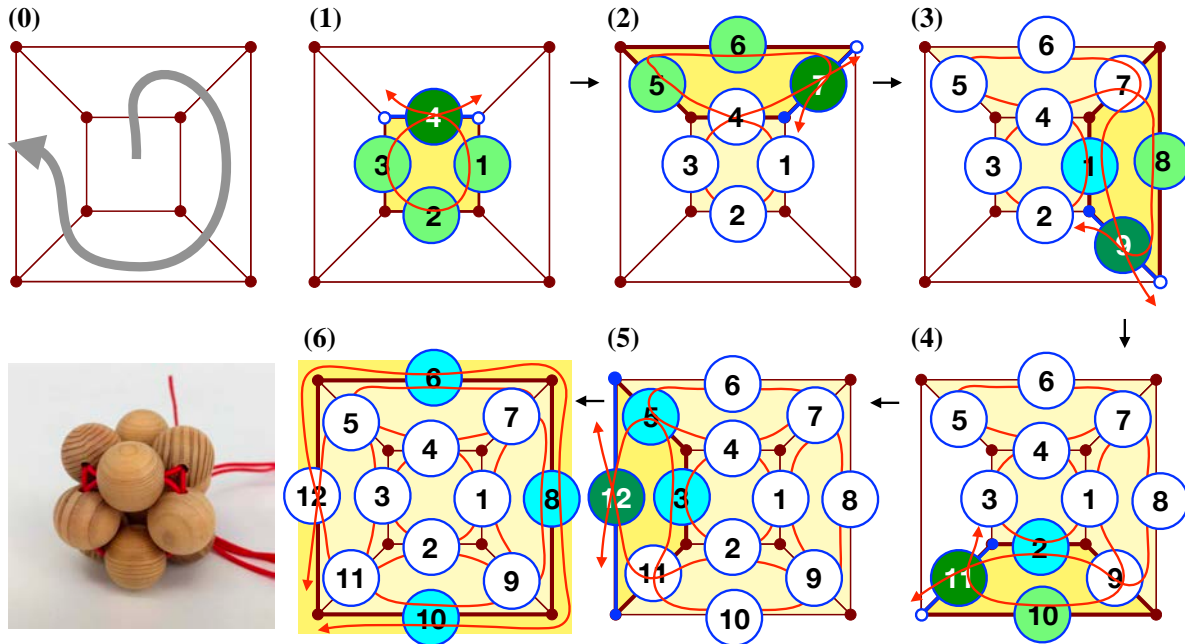


Figure 1: Constructing process of a beaded cube illustrated by using Schlegel diagrams. At each step, blue circles denote the beads just fished and green circles the beads just added. The dark green circle corresponds to the last bead just added and is labeled by the largest numeral. It is also the position where the crossing of two ends of thread take place and will be denoted as the *crossing bead*.

1. (a) String four beads (#1 ~ #4) in the middle of the thread.
 - (b) Take the right end and insert it through the furthest bead (#4) in the opposite direction and pull the thread tight. This makes the first square. For convenience, beads are labeled in ascending order according to the sequence they are added.
2. (a) Add three beads (#5 ~ #7) on the left end of the thread.
 - (b) Take the opposite end and insert it through the furthest bead (#7) in the opposite direction and pull the thread tight. This makes the second square. The first two rings look like a figure eight.
3. (a) In order for the next square to be connected with the two previous existing rings, the thread must go through bead #1 in the neighboring ring. Insert the end of the thread at the junction through the closest bead (#1)—this is called *fishing* and this end is denoted as the *fishing end* or *fishing line*. Notice that one will always add beads to the other end (*non-fishing end*) in the following step. Thus, the fishing and non-fishing ends will always alternate and both ends should decrease in length roughly equally.
 - (b) Add two beads on the non-fishing end.
 - (c) Take the other end and insert it through the furthest bead (#9) in the opposite direction and pull the lines tight. This makes the third square. This structure can be viewed as a hemisphere now.
4. (a) Go fishing (#2).
 - (b) Add two beads (#10, #11) on the non-fishing end.
 - (c) Take the other end and insert it through the furthest bead (#11) in the opposite direction and pull the thread tight. This makes the fourth square.
5. (a) Go fishing (#3, #5).
 - (b) Add the last bead (#12) on the non-fishing end.

(c) Take the other end and insert it through bead #12 in the opposite direction and pull the thread tight. This makes the fifth square.

6. Go fishing (through beads #6, #8, and #10) and complete the whole structure.

The whole procedure can be summarized in a tabular beading form as given in Table 1. At each step, decision should be made on which neighboring beads needs to be fished first, and then beads can be added to the non-fishing end in order to keep the beading following along the spiral direction. Although we usually view the spherical beads as edges of the corresponding polyhedron, one can also connect the center of neighboring beads and visualizes the bead model as a rectified polyhedron, in this case, a cuboctahedron. If we follow the RAW technique and keep attention to pulling tight two ends of the thread around the crossing bead at the end of each step, a robust beaded structure will be created automatically.

Step i	1	2	3	4	5	6
# of beads fished, f_i	0	0	1	1	2	3
# of beads added, n_i	4	3	2	2	1	0
# of beads in a ring, p_i	4	4	4	4	4	4

Table 1: Tabular beading form for creating a beaded cube. The last row corresponds to its spiral face-Hamiltonian path, which can be codified by the sequence “444444”.

According to the many workshops we have given, the most difficult step for beginners in constructing a beaded structure is *fishing*. The trick is to examine two ends of the thread that just coming out of the crossing bead every time one just finished a ring. Notice that the crossing bead at each step is always the last bead that has been just added! In Figure 1, a blue line segment is used to denote the channel direction of the crossing bead at the end of each step. The two ends of this segment correspond to two neighboring vertices of the degree three polyhedron. For instance, when the square 2 (the second step in Figure 1) is just complete, the particular end of the line that points to the direction of the solid blue circle must be the fishing line, while the other end pointing to the vertex denoted by empty blue circle has one edge without any bead on it must correspond to the non-fishing line. So the rule for deciding whether *fishing* should be performed or not is to check if the corresponding vertex (as indicated by the solid blue dot in the second step of Figure 1) is surrounded by three beads. If it is surrounded by three beads, go fishing, meaning that the cyan circle labeled by #1 as shown in the third step in Figure 1 should be fished before adding new beads.

The same beading method can be easily extended to other degree three polyhedron that contains a spiral face-Hamiltonian path, which visits each face once and only once along the spiral. For instance, a spiral face-Hamiltonian path exists for the great rhombicuboctahedron and can be codified as “8 46464646 84848484 64646464 8”. Without employing its Schlegel diagram or even the corresponding tabular beading form, this code already gives enough information for creating its bead model. One simply follows this code and start to construct a bead model by making one polygon after another either clockwise or counterclockwise. Special attention has to be paid to *fishing* before adding new beads at each step to ensure that the condition of a degree three polyhedron is satisfied. When the last ring is formed, an overall structure that gives a faithful three-dimensional bead representation for the corresponding polyhedron will be automatically created. In Figure 2, we show the bead models for four degree three polyhedra.

More generally, the algorithm for making any degree three polyhedron that contains a spiral face-Hamiltonian path specified by the code $p_1 p_2 \dots p_N$, where p_i is the number of sides of the i th polygon along the spiral, can be written as follows:

1. $i = 1$
 - (a) Add $n_1 (= p_1)$ beads to the string.

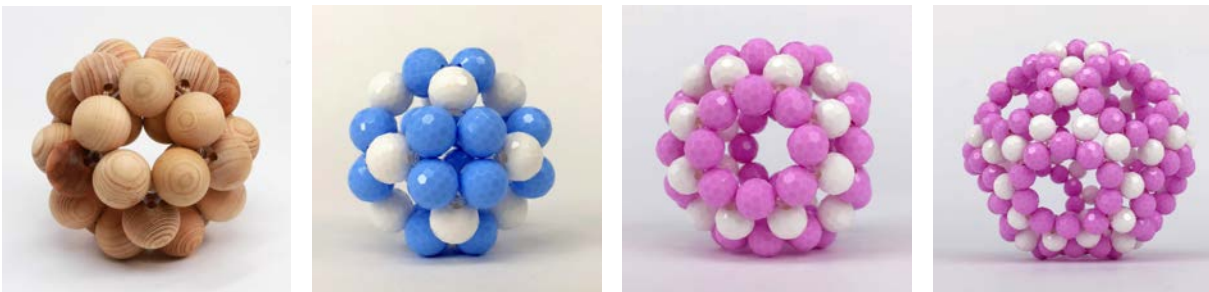


Figure 2: Bead models for dodecahedron, truncated octahedron, great rhombicuboctahedron, and great rhombicosidodecahedron.

- (b) Form a ring with p_1 beads by crossing both ends of the string at the crossing bead labeled by the largest numeral (i.e. the furthest bead or the last bead just added).
2. $i = i + 1$
 - (a) Fishing: $f_i = 0$, where f_i is the number of beads fished at the step i .
 - i. Fish the next bead if the corresponding vertex is saturated (or surrounded) by three beads, $f_i = f_i + 1$.
 - ii. Repeat 2(a)i until all neighboring saturated vertices are fished.
 - (b) Add $n_i = p_i - f_i - 1$ beads to the non-fishing end of the string.
 - (c) Form a ring that contains p_i beads (p_i -gon) by crossing both ends of the string at the bead labeled by the largest numeral.
3. Repeat 2 until all polygons are formed.

One can easily see that each bead is stitched exactly two times if the corresponding graph contains a face-Hamiltonian path. Therefore, one must choose the thickness of a thread such that it can go through each bead at least two times. However, without further handling of the remaining thread still outside the bead sculpture, the bead sculpture just finished can loosen up easily. The trick in preventing this happening is to string the remaining part of threads lying outside into the bead structure again until the whole sculpture becomes tightly bound. As a result, the channels of some beads will be filled by threads three or even four times.

Bead Sculptures: Curved graphitic structures

The first approach to the construction of bead models is based on the RAW techniques of mathematical beading introduced in previous section. According to the coordination numbers, there are three different broad families of nanostructures that can be constructed using the mathematical beading technique. The first family consists of any cage-like polyhedral fullerene, which is a degree three (cubic) polyhedral graph with only pentagonal and hexagonal faces, and the generalization to various curved graphitic structures that allow non-hexagonal rings other than pentagons. For simple cage-like polyhedral fullerenes C_v without holes (i.e. $g = 0$, where g is the genus), the number of pentagonal faces is always 12, the number of hexagonal faces is $(v - 20)/2$, and the number of edges (or beads) is $3v/2$. The bead model of a fullerene with v carbon atoms comprises exactly $3v/2$ beads since beads stand for chemical bonds instead of atoms.

To make any fullerene, weave face by face with beads using the RAW according to its pentagonal spiral code which specifies a spiral face-Hamiltonian path that visits each polygonal face once and only once. For instance, the spiral code for C_{60} , [1 7 9 11 13 15 18 20 22 24 26 32], gives the positions of twelve pentagons

in the spiral sequence of twelve pentagons and twenty hexagons. By using a prescribed spiral code, one can create a realistic bead model of any cage-like fullerene.

In addition to the cage-like fullerenes, beads can also be used to construct other more complicated graphitic structures such as carbon nanotori, high-genus fullerenes, and so forth. According to our experience, beads can be used to construct robust physical models of simple and complicated carbon nanostructures that are otherwise inaccessible. In Figure 3, we show a variety of curved graphitic structures which include finite and infinite 3-connected graphitic surfaces, starting from the simplest C_{60} to carbon nanotori, helically coiled carbon nanotubes, carbon torus knots, high-genus fullerenes, and triply periodic minimal surfaces with negative Gaussian curvatures. [11]



Figure 3: Curved graphitic structures. First row: Buckyball (C_{60}), endcapped carbon nanotube C_{80} , fullerene with lollipop shape, HIV virus, carbon nanotorus C_{120} ; Second row: curved nanotori, helically coiled carbon nanotube, carbon trefoil knot, dodecahedral high-genus fullerene, genus-3 carbon tetrahedron; third row: fused C_{60} cube, carbon tetrapod, single periodic minimal surface, I-WP surface, $(1, 0)$ -P-surface; Fourth row: $(1, 1)$ -P-surface, $(2, 1)$ -P-surface, Chen-Gackstatter surface, a genus-2 surface, intersection for four graphene sheets with four lines of sp^3 defects.

Bead Sculptures: Zeolites and tetravalent graphs

The second family that can be constructed by mathematical beading comprises tetravalent periodic structures with the degree (or valency) of a vertex equal to four, which include diamond, ices, clathrate hydrates,

zeolites, and many minerals. [12, 13, 14] In Figure 4, we give a few examples of bead models corresponding to various zeolite frameworks. Zeolites are porous aluminosilicates, widely used as efficient catalysts, adsorbents, and ion exchangers in petrochemical industries and in our daily life. Mathematicians have predicted that millions of hypothetical zeolite structures are possible. However, only 245 structures have been discovered and synthesized so far.



Figure 4: Zeolites specified by 3-letter Framework Type Codes. First row: SOD (Sodalite), LTA, FAU, EMT, RHO; Second row: KFI, AST, TSC, ATN, BCT; Third row: LTL, MOZ, SAS, ABW, BEC; Fourth row: MEP, MTN, DOH, MWW (MCM-22), SSF.

Zeolites are based on the primary tetrahedral building unit, TO_4 , where the central tetrahedrally bonded atom, T, is usually either a silicon or aluminum cation, surrounded by four oxygen anions. By linking these tetrahedra together, it is possible to build larger building units such as truncated octahedra, dodecahedra, etc. When these building units are linked together to form a zeolite framework, they characteristically yield periodic cavities and channels throughout the structure. It is a challenging work to construct all these structures with beads since the beading is an inherently slow sequential process. Right now, we have succeeded in making bead models for more than forty zeolite framework types.

Bead Sculptures: Inorganic structures with valencies higher than four

We have also applied successfully the technique of mathematical beading to the construction of skeletal polyhedral models with the degree of a vertex higher than four for many nanoscale inorganic structures or macroscopic octet-truss systems by using long tubular bugle beads. [7, 8] If bugle beads with the same length are adopted, all eight convex deltahedra can be constructed and the resulting structures are rigid as a consequence of the Cauchy rigidity theorem. Among these convex deltahedra, tetrahedron and octahedron are two most common structural motifs occurring in the molecular transition metal chemistry, inorganic solids, and many mineral structures.

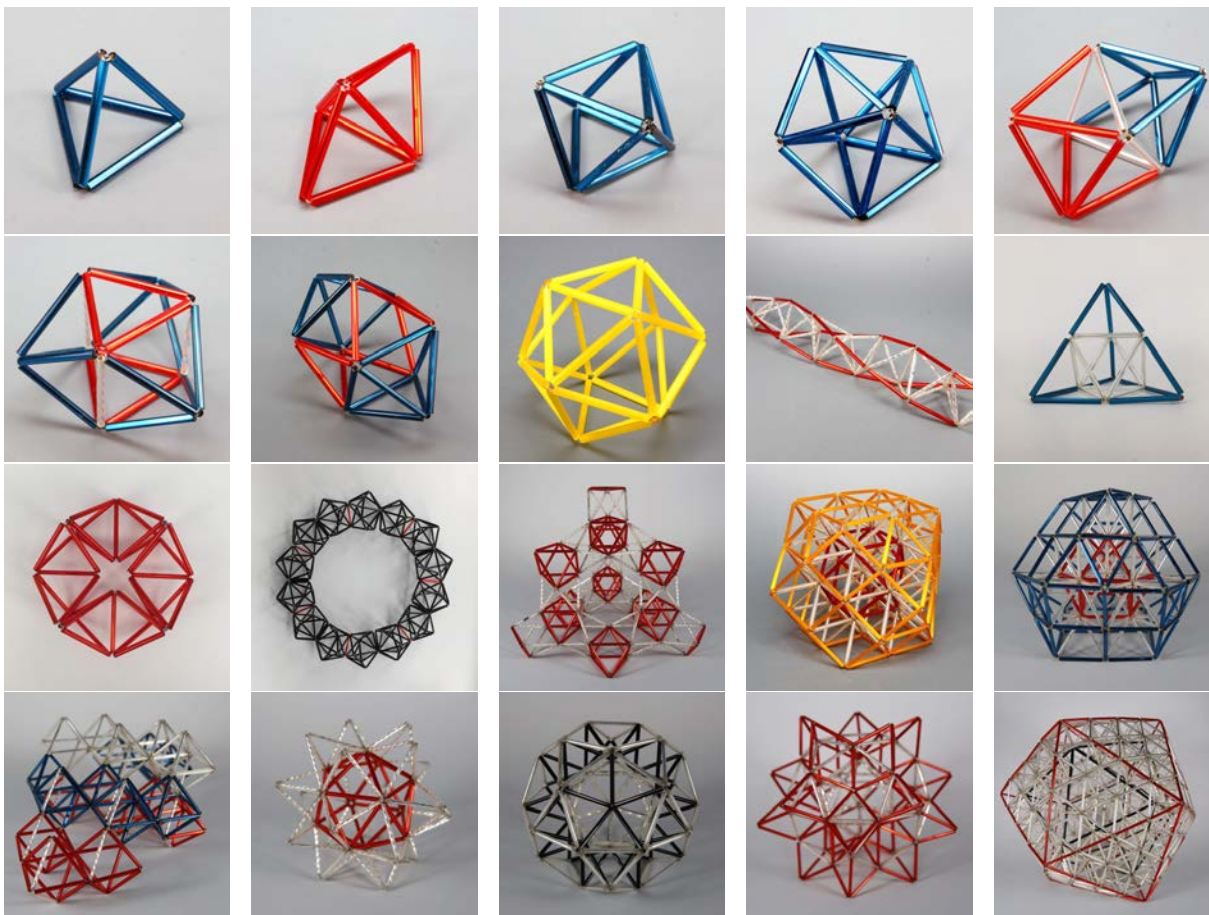


Figure 5: Skeletal truss models. First row: tetrahedron, trigonal bipyramid, octahedron, pentagonal bipyramid, snub disphenoid; Second row: Triaugmented triangular prism, gyroelongated square bipyramid, Icosahedron, Boerdijk-Coxeter helix; Third row: Kaleidocycle with eight regular tetrahedra, and Kaleidocycle with 14 gyroelongated square bipyramids, octet-truss consisting solely of octahedra, hexagonal close packed octet truss, cubic close packed octet truss; Fourth row: Anatase TiO_2 structure, elevated icosahedron, compound of icosahedron and 20 octahedra, compound of icosahedron and rhombic hexecontahedron, frequency-4 Mackay icosahedron.

Other deltahedra can sometimes become important in nano world. For instance, the B_{12} motifs in the extended structure of α -rhombohedral boron are found to be icosahedra. Many larger molecular structures can be described by linked deltahedra through sharing common vertices, edges, or faces. If bugle beads with different lengths are used, a richer structural variety of skeletal polyhedral sculptures can also be built.

Figure 5 shows a few skeletal truss models for inorganic systems and space frameworks based on linking different numbers of deltahedra.

Bead-Chain Construction Set and Interlocking Puzzles

Making a bead model for a large molecule is inherently a slow sequential process, and can take a lot of time to complete except for certain modularizable systems which could be constructed by many people together. In this section, we show that it is possible to create the same valence sphere models without any beading, at least for certain molecules such as polycyclic cage-like hydrocarbon with the chemical formula $C_{2n}H_{2n}$, or polyhedrane. Starting from several pre-made building blocks, the valence sphere models for these polyhedrane molecules can be assembled through cross-linkings and the tedious beading procedure can be avoided completely.

Consider the simplest polyhedrane, a cubane molecule with the formula C_8H_8 . This molecule consists of eight carbon atoms arranged at the corners of a cube, with one hydrogen atom attached to each carbon atom. [15] Also there are twelve CC single bonds and eight CH bonds in this molecule. Notice that the bead model for this molecule corresponds to its valence sphere packing model. Atoms are not explicitly shown, while twenty chemical bonds are represented by twenty beads. To make a bead model for this molecule with a finite number of linear bead chains through cross-linkings, the beads corresponding to eight CH bonds must be located at chain ends. Hence, exactly four bead chains are required in order that a chain can start from one CH bond and end at another CH bond. Hence, CH bonds are represented by terminal beads. To use as few basic building blocks as possible, we choose $12/4 = 3$ non-terminal beads for each chain. Thus, a single type of five-bead chain is sufficient for making a bead model of a cubane molecule. Interestingly, the assembly process of a cubane model from four five-bead chains is nontrivial since the many possible ways of making cross-linkings among four five-bead chains exist. In many ways, assembling the bead model of a cubane molecule bears a resemblance to solving an interlocking puzzle. Therefore, we call this model as the BC-Cube (Bead-Chain Cube) puzzle as shown in Figure 6 and 7a.

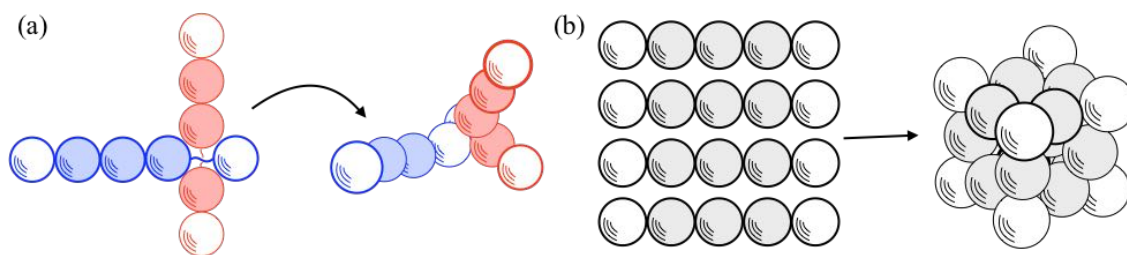


Figure 6: (a) Cross linking of two bead 5-bead chains. (b) A BC-Cube can be assembled from four five-bead chains through eight cross linkings.

Figures 7b and 7c shows the bead-chain models of the other two Platonic hydrocarbon molecules, namely tetrahedrane and dodecahedrane, which can be assembled from two and ten five-bead chains, respectively. Bead-chain models for two of seven Archimedean polyhedranes, truncated tetrahedrane ($C_{12}H_{12}$) and truncated icosahedrane ($C_{60}H_{60}$), are shown in Figures 7d and 7e, respectively. More generally, the carbon skeletons of polycyclic hydrocarbon with formula $C_{2n}H_{2n}$ can be regarded as cubic graphs in which all vertices have degree three. If we ignore CH bonds, each carbon atom is bonded exactly to three neighboring carbon atoms. A cross-linking between two bead chains introduces a local tetrahedral arrangement for four beads. To satisfy the requirement that carbon atoms are located on the polyhedral skeleton and hydrogen atoms pointing outward, one needs exactly n five-bead chains for making a bead model of the corresponding polyhedrane molecule. Finding a procedure of making a bead-chain model for a polyhedrane with five-bead

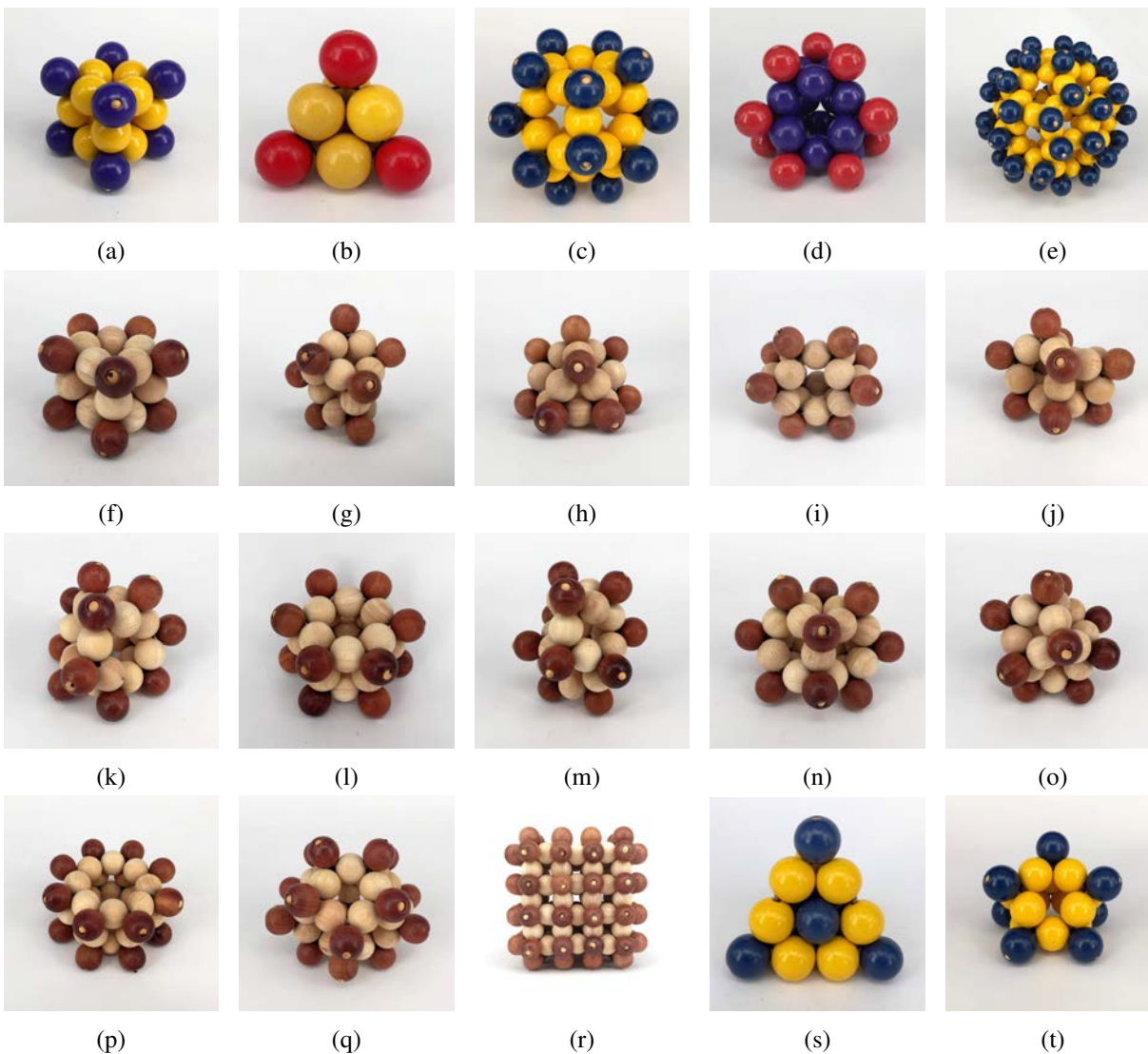


Figure 7: Bead chain construction set. First row: BC-Cube (C_8H_8) with four five-bead chains, BC-Tetrahedron (C_4H_4) with two five-bead chains, BC-Dodecahedron ($C_{20}H_{20}$) with 10 five-bead chains, BC-truncated tetrahedron ($C_{12}H_{12}$) with 6 five-bead chains, BC-Buckyball ($C_{60}H_{60}$) with 30 five-bead chains; Second row: fiver isomers of $C_{10}H_{10}$; Third row: five isomers of $C_{12}H_{12}$; Fourth row: $C_{16}H_{16}$, $C_{18}H_{18}$, Perovskite structure constructed by 48 five-bead chains, close-packed tetrahedral arrangement of twenty spheres, another isomer of C_8H_8 that contains two triangular faces and two pentagonal faces.

chains is equivalent to finding a way to draw its skeletal polyhedron with n non-overlapping strokes and each stroke covers exactly three edges.

Since the total number of cubic graphs grows exponentially as a function of n , there is a rich variety of possible structures that can be constructed by this new kind of bead chain construction set. In Table 2, we list the number of unlabeled cubic and cubic polyhedral graphs up to 22 vertices. Whenever an n -stroke solution for a polyhedrane exists, the corresponding polyhedral bead-chain model can be constructed.

Table 2: Number of unlabeled cubic polyhedral and triangle-free cubic polyhedral graphs with $2n$ nodes, with $n = 2, 3, \dots, 11$.

Vertex	4	6	8	10	12	14	16	18	20	22
Cubic	1	2	5	19	85	509	4060	41301	510489	7319447
Polyhedral	1	1	2	5	14	50	233	1249	7595	49566

Moreover, if we allow cross-linkings of three bead chains at a single position, more complicated nanostructures that contain local octahedral arrangements of six spheres can also be created. For instance, Figure 7r gives a bead model for a portion of the perovskite structure that contains $4 \times 4 \times 4 = 64$ unit cells. A total of 48 five-bead chains with 64 triple-chain crossings are used. Finally, notice that the same four five-bead chains, which we used for making the bead model of a cubane molecule, can also be used to make another two interesting structures, one is a close-packed tetrahedral arrangement of twenty spheres (Figure 7s) and another one corresponds to a different isomer of C_8H_8 (Figure 7t)! We leave this as a puzzle for readers.

Conclusion

In conclusion, chemistry is rich with many interesting structures which can be employed for constructing aesthetically pleasing bead sculptures. With suitable interpretations for the meaning of beads and strings, these sculptures can model many microscopic molecular structures faithfully. More importantly, we believe that the molecular and structural diversities at the nanometer scale can also give us inspirations for finding new forms and shapes of sculptures and architectures.

Two different approaches for constructing bead sculptures, one by the mathematical beading and the other through the bead-chain cross-linking, are presented in this paper. The first approach based on mathematical beading can be applied to a large variety of nano structures to create fascinating bead sculptures. The disadvantage is that the mathematical beading is inherently a slow and sequential process. Great patience is required if one wishes to construct a bead sculpture with thousands of beads. The second approach based on the cross-linking of pre-made bead chains involves no beading at all. We demonstrate that the valence sphere model for any polycyclic hydrocarbon molecule of general formula $C_{2n}H_{2n}$ with carbon atoms located on the nodes of corresponding cubic graph can be assembled from n linear five-bead chains through n cross-linkings, which can be regarded as a new type of interlocking puzzle.

Acknowledgements

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