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The Architecture of Two-Dimensional Origami Links

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Abstract: As a new species of links, the two-dimensional origami links have been constructed by our method of "tangles covering". In order to facilitate the architecture of DNA origami links, tangles have been substituted by 4 degree vertices. Then a new term of links has been defined as "boundary". After that, the relation between the twist- number of tangle and the components of the links has been analyzed. At the end, a 2-regular graph has been established in a connected shape with random boundary. It was proved that the shape could be synthesized by DNA origami method in theory. This study may extend the content of knot theory and the application of DNA origami in experiments.

Introduction

Due to its special structure with two helical chains of nucleotides held together by the specific hydrogen-bonded base pairs, DNA has always been utilized as an important and ideal building material in creating programmable and predictive supramolecular structure [1]. In 1994, Adleman et al. reported that searching Hamilton circle in molecular design by computer experiments [2]. During the past few decades, various DNA origami links or catenanes [3] (e.g., DNA planar structures, rectangles, five-point stars and smile faces) have been synthesized in Rothemund's lab which open the door of DNA origami era. Then, more regular and irregular, two dimensional and three dimensional DNA origami styles occurred, for example, two dimensional arrays[4], printed origami structures[5], origami Jigsaw Pieces[6], tetrahedron[7], cube[8-9] and Möbius surfaces[10].These structures are so interested that both chemists and mathematicians are attracted to focus attention on them because of their significantly geometrical characters and potential application.

With these exciting and intriguing results in depth, Qiu's group has devoted considerable effort to describing some of these amazing structures from the geometrical and topological points of view, which is based on knot theory [11-25].

Although the experiments and theoretical research about DNA origami is developing greatly, there are still many issues are undetermined. The first problem is that can any connected graph be created by DNA origami. Second, components of links corresponding to DNA strands are circles inevitably, but there are many flexible ends in structures that created by DNA origami. So it is worth to consider how to deal with these flexible ends.

The two problems inspire our great interest in concerning about the construction of DNA 2D origami links theoretically. Therefore, the aim of our paper is to prove that any connected graph can be created by DNA origami; we construct two-dimensional origami links by our method "tangles covering" which solve the second problem. These studies give chemists and biologists a new access to checkout and develop new synthesis strategies.

Constructing planar links by DNA origami

The tangle model in knot theory is used to construct DNA origami links. Firstly, an unoriented tangle (**UT**) acting as the building blocks is defined as follows:

Definition: Given a projection of a knot or a link, the tangle **UT** is a region in the projection plane surrounded by a circle in which the knot or link crosses the boundary four times and, there are only two crossing lines inside, as depicted in Figure 1. There are two chiral tangles that are mirror-image, the half-twist number k ($k \geq 0$, and k is nonnegative integer) in these simple tangles is the same as the crossing number. Meanwhile, an axis is defined which goes along with the increasing half-twist number.

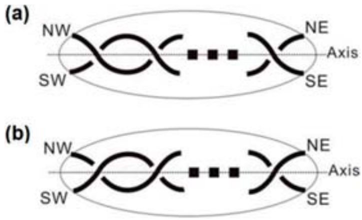

Figure 1. The two mirror-image simple **UT**, (a) right-handed; (b) left-handed.
The four ends of tangles are labeled as NW, NE, SW and SE, respectively.

Since the UT is defined on a projection region, all operations should be realized on the projection, so the UT only has a twofold-rotational symmetric axis vertical to the projection. The tangle in Figure 1a is selected as the building blocks because the two mirror-image tangles have the identical constructing methods.

The constructing processes should abide by the rules as below: the four ends of the tangles cannot be connected together with each other. Otherwise, a trivial knot will be produced. However, the ends on the same side of the axes can be connected together, and a two-stranded link occurs if the half-twists number is even, while a k_1 knot occurs if the half-twists number is odd.

The interior and exterior of the graph is quite different during the constructing processes. The boundaries are composed of flexible ends whereas it is not for the interior. Herein, the interior of the graph is discussed firstly. At the beginning of the motif growth, the tangle is considered as a rigid motif. There are six connection types of flexible ends:

{NW-NE};{NW-SE};{NW-SW};{SW-NE};{SW-SE} ;{NE-SE}

But only the types {NW-SE} and {SW-NE} satisfy the interior of the graph. It is obvious that there are two increasing directions when the tangles are connected together, one is vertical to and the other is parallel to the axes, as shown in Figure 2a. The growing of a graph results in a motif having a twofold-rotational symmetric axis. The building blocks determine the symmetry of the resultant motif. Of course, it is possible that there appear the forbidden crossings in the processes. It seems to represent the two connection models, so it is necessary to discuss the connecting relationship between the tangles. To simplify, the tangles are transformed into a 4-degree points, and then the question is about the relationship referring to the connections of the 4-degree points (Figure 3), which can be resolved by graph theory.

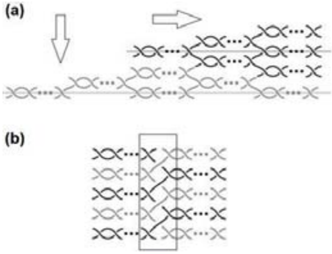

Figure 2. Connection types of tangles. (a) two increasing directions; (b) emerge forbidden crossings.

The same method is used to deal with the type presented in Figure 2b, so the forbidden crossings is converted into the special 4-degree points, as shown in Figure 4a, and thus the connection between the tangles are transformed into the 4-degree points. The over and under crossings represented by the 4-degree points are converted, as stated below: starting from any point, change all the points on the paths that the start point placed over (or under), as described in Figure 4b. Then the paths form a network (Figure 4c). Coloring the points in Figure 4c with other color, it is surprising that the type in Figure 3b is obtained. This operation indicates that the forbidden crossings cannot change the connection between the tangles. In other words, the relationship between tangles has the two different connection styles. So the interior of graph can be constructed by this way, it satisfies the partial characters of bipartite graph.

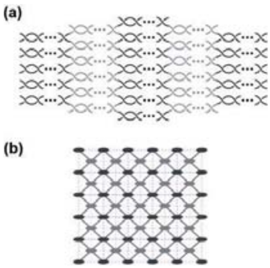

Figure3. Connection between tangles is transformed into connection between 4-degree points (no forbidden crossings). (a) tangles are joined which marked with gray and black, the same color are not joined; (b) the gray full lines indicates the relation between 4-degree points, points with same color array lattices.

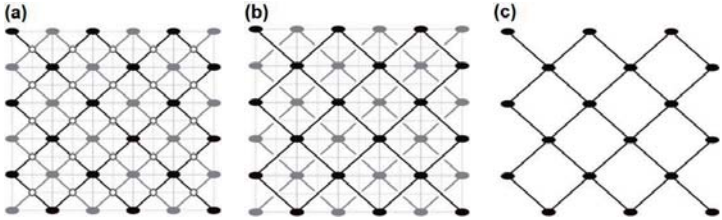

Figure 4. Connections with and without forbidden crossings.

With respect to the tangles mentioned above, there are three types of the 4-degree point on the boundaries, in which the number of the flexibly ends can be 1, 2 or 3. When the number of the flexibly ends is 1 or 3, the sum of the number of boundaries points must be even (including 0). We can prove this conclusion as below:

The number of the flexibly ends of a 4-degree point is 4, so the total number of flexibly ends is the multiple of 4, that is to say it is even. When the 4-degree point is connected by the flexibly ends, then the number of flexibly ends is reduced by 2 or its multiple. The final number of the flexibly ends should be even, then cut off the points with 2 flexibly ends, it is still even, indicates as M which could be written as $1m+3n$. So $m + n$ must be even because M is even.

The flexibly ends are processed as follows: no circle self-assembled by 4-degree point tangle on the rule as we define for tangles connection. So the process can be separated into two parts bases on the characters of flexibly ends. The first part is all boundaries points with 2 flexibly ends, then any adjacent points join together in turn and cannot assemble into a circle, so 4-regular graphs are obtained; the second part is that some of boundaries points with odd flexibly ends, we need to deal with these points by two ways further. One is cut off them which is defined below:

1. If the boundary point has 3 flexibly ends, cut it off, the number of the flexibly ends for the point connecting to it increases 1. If the flexibly ends are larger than 2, then repeat the above operation. The number of the flexibly ends for the other boundary points 1and 2 remain even.

2. Starting from a point with 1 flexibly end, joining flexibly ends along clockwise or anti-clockwise until reach to another point with 1 flexibly end. All points with 1 flexibly end and the portion points with 2 flexibly ends will be transformed into 4-degree points by this method.
3. For the other points with 2 flexibly ends, replace them with arcs, then 4-regular graphs are realized.

The other exists a danger that cut off points with 3 flexibly ends; only a 4-degree point will appear. Then make these points assembled into circles (transform points into tangles will be discussed in next section), use the steps 2 and 3 to deal with the signal flexibly end, a 4-regular graph is obtained at last. Transforming the points of 4-regular graph into tangles, then origami links is realized.

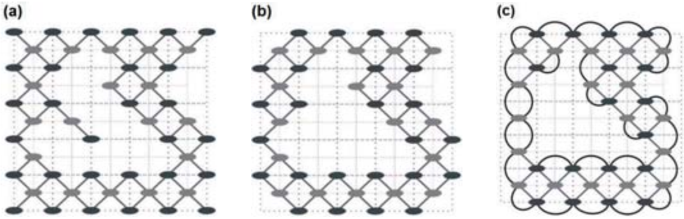

Figure 5. Boundary treatments. (a) construct interior as boundary treatment; (b) cut off points with 3 flexibly ends；(c) deal with other flexibly ends.

Analysis the strand paths of origami links

Kuzuya and Komiyama have discussed the influence of the odd or even number of DNA half-turn on the scaffold motif and on the short strands with “S” or “Z” shape [26]. Here we will discuss the relationship between the number of half-turns and links in detail. The number of half-turns（$k \geq 0$）determines the relationship between the two strands in tangles and the four flexibly ends {NW; NE; SW; SE} (shown in Figure 6). Figure 6a shows the even number of half-turns, meaning two flexibly ends of strand on the same side, while Figure 6b is the odd number of half-turns having two flexibly ends of strand on the opposite side.

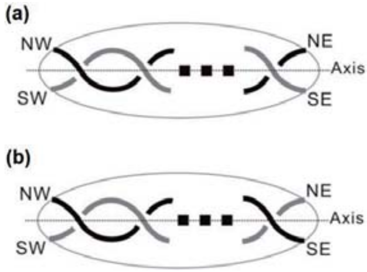

Figure 6. The number of half-turns *k* influence on tangles,
(a) even half-turns; (b) odd half-turns.

For the case with even tangles, a special case appears when $k=0$, as shown in Figure 7. The tangles in Figure 7a and b are still even tangles based on half-turns, though they are defined as 0 tangle and ∞ tangle, respectively. Using the constructing method mentioned above, the final results are obtained (Figure 7c and d), they are trivial knots. A trivial knot will occur if the tangles in Figure 7a and b are combined together.

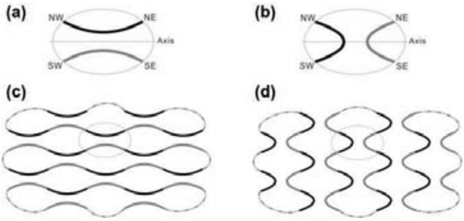

Figure 7. Tangles with the number of half-turns *k* is 0. (a) 0 tangle; (b) ∞ tangle; (c) Join 0 tangles together; (d) join ∞ tangles together. Boundary treatment are colored by gray double dots lines.

If $k > 0$, there are two different types for the joint tangles as plotted in Figure 8. After dealing with the boundaries, there gets a knot (Figure 8a), while a link that strands self-wind and nest to each other is obtained (Figure 8b). It can be seen that starting from any tangle on boundaries along the path that one of the tangle strands passes and ends at any tangle on boundaries, the even or odd number of tangles determines assembling directions and thus the number of strands. The directions of paths and axes of tangles are parallel. If the numbers of tangles of paths are the same, the strands number are 1 when they are all even, parity alternate with each other or all odd are more than 1.

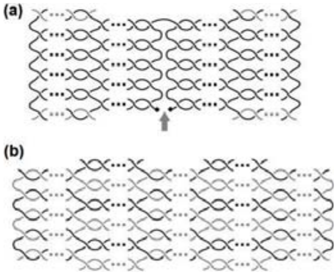

Figure 8. Different paths of even tangles (k>0).
(a) a knot; (b) a link that strands self-wind and nest to each other.

With respect to the case with all odd tangles, if k=1, it is a cross, then a special case occurs as shown in Figure 9. If the direction is not decided, tangles in Figure 9a and b are identical. Using all tangles in Figure 9a or b to construct, and the parallel lines that over and under layer cross together rather than networks are obtained. Networks in Figure 9c is realized by joining tangles in Figure 9a and b alternately.

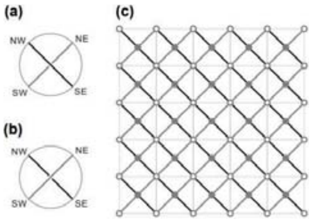

Figure 9. Half-twist numbers of the tangles are satisfied by k=1, (a) and (b) join together into network (c), circles and gray points indicates (a) and (b), respectively.

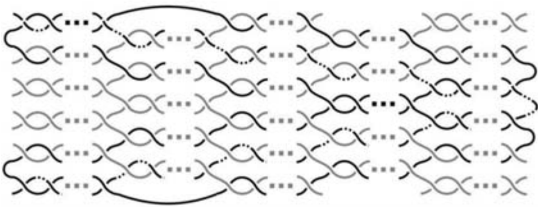

Figure 10. Structure with k>1 odd tangles. The double dots line and black full line indicate paths that deal with different methods.

However, the structure constructed with $k > 1$ is shown in Figure 10. The double dotted lines show that starting from any tangle on boundaries and along one path that travels the tangle and ends the next boundaries. In this way, a "Z" shape strand that travels the entire graph is realized. In the final links, the "Z" shape strand is assembled into a circle (Figure10, the "Z" strand denoted by black solid line is essentially different from the "Z" of short strands reported by Kuzuya [26]). There is a fixed angle between the direction of path and axes of tangles. The final link is composed of such circles nests together.

In the following section, the case constructed with even and odd tangles are discussed. There are three cases base on different construction methods. For the first one, the interior and boundary of graph are decorated with all odd tangles and some even tangles, as displayed in Figure 11a and b. The second one is characteristic of all even tangles inside and some odd tangles on boundary (Figure 11c and d). The third one is composed of odd and even tangles travelling graph alternately (Figure 11e and f). A definition is illustrated here for the discussion.

Definition: If one path starts from one tangle on boundary and travels one of two strands, then ends at the next boundary, give a power **1** to the traveling path.

By analysis, we can discover that although the structures shown in figure 11a, c, e are different, they have one thing in common, the number of tangles travel on the path with power 1 is even, the power of the path that travel tangles is an integer more than 2. However, the circle paths in figure 11b, d, f with power 2 and the numbers of tangles that every path with power 1 travels are odd.

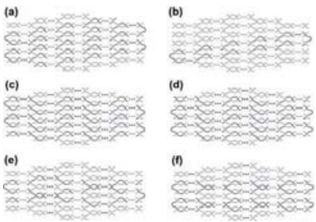

Figure 11. Odd and even tangles coexist in one graph. Even tangles are indicates by black points. a,b: the main part is odd tangles, boundary is decorated with even tangles; c,d: the main part is even tangles, boundary is decorated with odd tangles; e, f: odd and even tangles travel graph, alternately.

Nevertheless, the differences among the structures shown in Figure 11a, c and e are obvious. There is a complementary path of the structures shown in Figure 11a, c respectively because the construct results of structures are paths that travel all tangles once as indicated by the black lines. The connection between two paths determines the number of strands. A link composed of two strands is produced if the tail and head are joined together, and there is a knot with two strands joined together. But the structure in Figure 11e needs two strands that each travels some points twice. Similarly, there is a link with two strands or a knot. It is similar to the structure shown in Figure 8a. For constructions shown in Figure 11b, d and f, the numbers of strands are more than 2, which are similar to constructions shown in Figure 8b and Figure 10.

The six unexpected cases for the flexibly ends connected

The points with 3 flexibly ends assemble themselves into circles, but when they are converted into tangles, some problems as shown in Figure 12 are derived. Firstly, a simple definition of Reidemeister operation is given.

Reidemeister operation I: A crossing is added to or removed from as represented in Figure 12a.

Reidemeister operation II: Two crossings are added to or removed from as represented in Figure 12b.

In Figure 12c and d, the flexibly ends distributing on two sides of axes are connected together. They can be replaced by an arc, and the strand number is 1. However, the flexibly ends on the same side of axes are connected together with different number of strands because of the various numbers of half-turns. The strands number of even tangles is 2, while it is 1 for odd tangles in which the Reidemeister operation is forbidden. It shows that the Reidemeister operations do not change the connection of flexibly ends, no matter what the strands number is. This is a powerful proof that our treatments on interior and boundary with different connections of flexibly ends are reasonable.

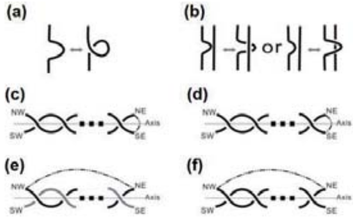

Figure 12. Flexibly ends of one tangle join together.
a: Reidemeister Ⅰoperation; b, d indicate connection of even tangles; c, e of odd tangles.
Double dots lines show treatments of flexibly ends of boundaries.

It is possible that two tangles {NE-NW} and {SE-SW} can be joined together simultaneously. The number of the half-turns increases correspondingly if tangles with the same chirality, and two uncrossed lines appear if two tangles are mirror-image.

Construction of mirror-image tangles

Construction of mirror-image tangles has somewhat different from the methods in the preceding sections because the crossing points are alternately, as shown in Figure 13, the black and the gray are joined together. However, the crossing points are the same and not alternately if only the black tangles (or gray tangles) are joined together, as can be seen in Figure 13 with the same color.

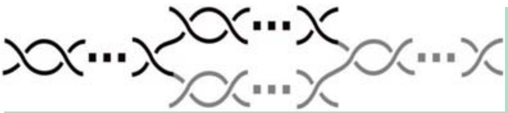

Figure 13. Connection of mirror-image tangles, black and gray tangles are mirrored.

Construction of orientated tangles

It is necessary to consider the orientation matching problem in discussing the connection of orientated tangles. For the constructions of orientated odd tangles (Figure 14a) or their mirror-image tangles (Figure 14c), their orientations match each other without considering the rotation operation. But with regard to the orientated even tangles (Figure 14b) or their mirror-image tangles (Figure 14d), their orientations do not match, so it needs the twofold rotational operation to make sure that the orientations match each other. The tangles that are mirror-image can be joined together directly because their orientations match.

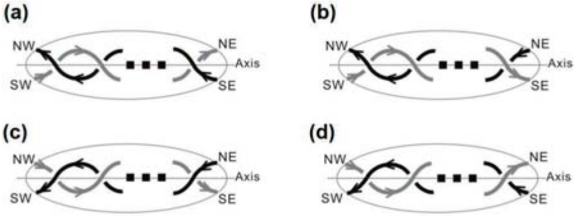

Figure 14. Orientated tangles. a, c are mirrored odd tangles; b, d are mirrored even tangles.

As a matter of fact, the links constructed by unoriented tangles have the similar construction methods and properties to those of the oriented links when we orient every strand based on the rules of oriented tangles.

The applicability of origami links

In this section, we will give a proof about the broader applicability of origami links to any connected graph with the help of bipartite graph. Given a connected region, plane, curved surface or three-dimensional space, it can be covered by a dot matrix (or a series of ordered points). Cut off the points outside of the given region. All the points in the region have the degree of 2, and a connected graph with a single strand joined by these points is obtained, so the graph is a 2-regular graph.

Definition: Graph G is a connected graph when any two points are joined by one path.

Theorem: Graph G is an Euler graph if and only if G is a connected graph that all degrees of vertexes are even. The Euler circle travels all edges once.

According to the theorem, the 2-regular graph described above possesses an Euler circle. The circle travels not only every edge once, but also every point once because the degree of each point is 2. Therefore, the Euler circle is a Hamilton circle. Transforming the Hamilton circle into loop and extending the degree of all points from 2 to 4, then a Hamilton graph is produced. Constructing origami links of whole connected region is completed if the 4 degree points are replaced by tangles. As a result, we can summarize that any connected graph could be constructed by origami links no matter what connected region is. Plane, curved surface and three-dimensional space satisfy. There are many Hamilton circles of a given connected region. In the following section, a simple discussion about planar graph is illustrated to describe the relationship between the links derived from these Hamilton circles.

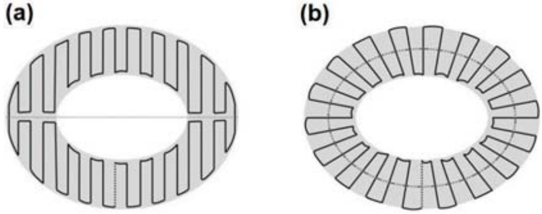

Figure 15. Hamilton circles of a given connected region.

As can be seen from Figure 15, the black line indicates path that travels the whole planar area, which is connected by the dotted line to form a Hamilton circle. There is a seam (as shown in Figure 15a indicates by dotted line) occurred if the path travels as the scaffolds do in experiment. The seam is rooted in one of the properties of Hamilton circle that without self-intersections. There is also a Hamilton circle that travels the whole region in Figure 15b. A double dotted line intersects with the Hamilton circle, a link will be realized if the points of intersection are replaced by tangles. To make a visualized distinction between these two links shown in Figure 15a and b is to seek a seam.

Conclusion

In this paper, we made a few attempts to design and study DNA origami links by mathematical methods. A series of DNA origami links were constructed by our method "tangle covering", which based on graph and knot theory.

The origami links have been realized successfully with connecting tangles together. We discovered that the boundaries number that with 1 or 3 flexibly ends must be even, and then we proposed the way that transforms flexibly ends into 4-degree vertices. We analysis the strand paths of DNA origami links of odd tangles links and even tangles links, respectively. At the same time, the links that are made of odd and even tangles alternatively are discussed. The laws that they assemble into links are put forward.

Finally, the six unexpected cases for the flexibly ends connected, construct mirrored tangles and orientated tangles are also investigated here. In a word, we have studied that construct DNA origami links for any cases and proved that any connected graph can be realized by the strategy of DNA origami. The methods reported here can supply theoretical basis for DNA experiments and its extension fields.

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