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The Architecture of DNA Sierpinski Links

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Abstract: For understanding the growth mechanisms of DNA fractal links, we constructed a series of Sierpinski links with different fractal number *n* based on *k*-tangle models and deduced the component number of them from some formulas. The properties of the links are investigated and DNA strand models with one three-fold symmetry axis are given. Moreover, a bottom-up synthesis method is proposed and extended to three dimensional DNA Sierpinski structures. Our results not only exhibit novel geometry but also provide a theoretical basis for synthesis of DNA fractal nanostructures.

1. Introduction

The special structure of DNA, that is, two helical chains of nucleotides held together by the specific hydrogen-bonded base pairs, render it as an important and ideal building material in creating programmable and predictive supramolecular structure [1]. During the past few decades, various DNA supramolecular structures including DNA polyhedral links or catenanes [2,3] (e.g., DNA tetrahedron [4–10], DNA cube [11–14], DNA truncated octahedron [14], DNA octahedron [15–17], DNA dodecahedron [6,18], DNA icosahedron [19,20], DNA bipyramid [21], and DNA buckyballs[6]) and others[22–28] have been synthesized by the experiments[29]. These structures are of great interest to both chemists and mathematicians because of the significantly geometrical characters associated with polyhedra [1, 29].

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Accompanied by these exciting and intriguing results, considerable effort has been devoted to describing some of these amazing structures from the geometrical and topological points of view, which is based on the knot theory [30, 31]. In recent years, Qiu's group has proposed the methods of "*N-branches curves and M-double-twisted lines covering"* and "*N-crossing curves and M-double-twisted lines covering"* to construct polyhedral links [1, 29, 32–39], especially for a series of surprising DNA polyhedral links [37–39]. At the same time, mathematical characterizations of these structures using topology and graph theory as working tools are making great progress [40–49]. More recently, Rothemund et al. have found that DNA can be assembled into crystals with patterns of Sierpinski triangles [50]. This importantly experimental result has inspires our great interest in concerning about the construction of DNA Sierpinski triangles theoretically.

In this paper, the aim is to design more perfect DNA Sierpinski triangles and to unravel the growth mechanisms of DNA Sierpinski links. In order to do these, DNA Sierpinski links are constructed with different fractal number n and, the growth mechanisms are studied. Furthermore, a bottom-up synthesis method of DNA Sierpinski triangles is put forward and extended to 3D model. These mathematic models may help chemists and biologists to checkout and develop new synthesis strategies, and thus enrich the system information of DNA nanostructures.

2. Construction of DNA Sierpinski links

The Sierpinski triangle, namely, the Sierpinski gasket or the Sierpinski Sieve, is a fractal and attractive fixed set described by Sierpinski in the following three steps [51]:

(i) Begin with an equilateral triangle;

(ii) Shrink the triangle to 1/2 height and 1/2 width, make three copies, and place the three shrunken triangles so that each triangle connects the other two triangles at a corner;

(iii) Repeat step (ii) with each of the smaller triangles.

As shown in Figure 1, a series of neoteric and fractal structures occur finally. Labeling them as E_0, E_1, E_2, E_3,..., E_n in order, it can be found that they conform to the law: one grows into three, three grow into nine..., so three E_{n-1} can grow into E_n, and the joint points are defined as growing points.

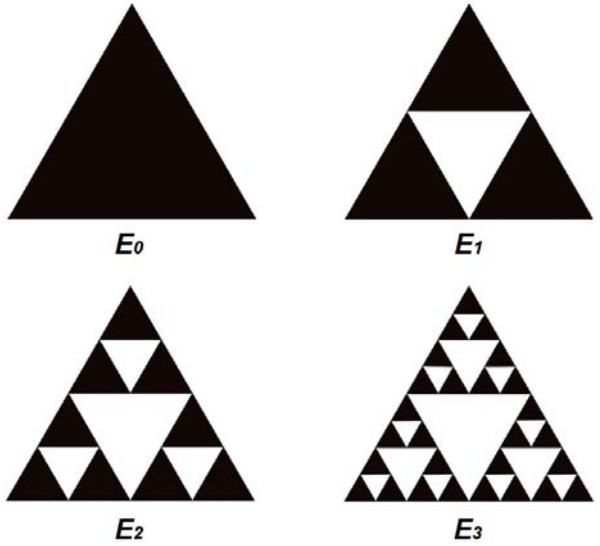

Figure 1. Sierpinski triangles with different fractal number *n*.

Two building blocks should be considered and designed in terms of the "*N-branched curves and M-double-twisted lines covering*" method. One is "*N-branched curves*", which are designed to cover the vertexes. The other is "*M-double-twisted lines*", which are used to replace the edges. And the combination of these two would lead to some closed circles. According to Sierpinski triangles, tangles [31] and two styles of vertexes [52] are constructed to fulfill the requirements. Three types of tangles (*i. e.* odd tangle, even tangle and the special case of zero tangle) are constructed on the basis of the twist number *k*, as represented in Figure 2. By covering the vertex with *n*-degree vertex (where *n* indicates the degree of vertex), two styles of three-degree vertexes are obtained in which one is crossed and the other is uncrossed (see Figure 3).

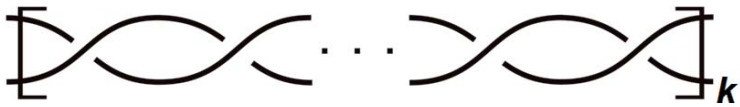

Figure 2. Tangle with *k* crossings.

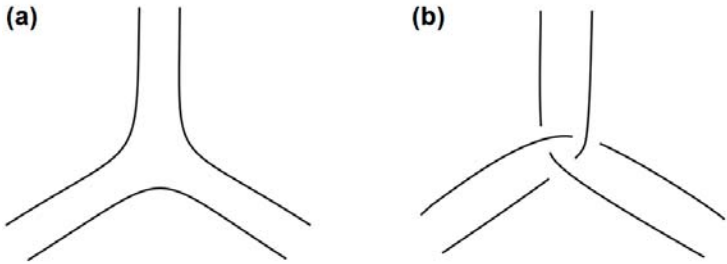

Figure 3. Two types of three-degree vertex, (a) the uncrossed three-degree vertex; and (b) the crossed three-degree vertex.

The links constructed are classified into two types depending on the styles of the growing points (crossed or uncrossed). They are the first type of DNA Sierpinski links when the growing points are crossed; otherwise, they are the second type. Three types of links (odd tangle, even tangle and zero tangle links) need to be constructed because the twist number k determines the strand number of link. For convenience, the cases of $k= 0, 1, 2$ were discussed in the following sections because the component numbers of them are the same as that of zero, odd and even tangle links.

2.1 The first type of DNA Sierpinski links

(i) DNA Sierpinski links with odd tangles

Using odd tangles to cover Sierpinski triangle E_n, the corresponding link labeled as $L_{o\text{-}n}$ is obtained, in which n is the fractal number. Figure 4 shows the Sierpinski links covered by $k=1$ tangles and uncrossed vertexes, and n is in the range from 0 to 4. It is evident that a trivial knot is derived if n=0. Moreover, the growth of links has an intrinsic mechanism when n increases. Also can be readily seen is that $L_{o\text{-}0}$ has 1 DNA strand, while $L_{o\text{-}1}$, $L_{o\text{-}2}$ and $L_{o\text{-}3}$ have 2, 5 and 14, respectively. The number of DNA strands is defined as the component number, denoted as A_n. Thus, the growing mechanism can be illustrated as the difference between A_n and $A_{n\text{-}1}$ which equals to 3^{n-1} during the assembly of odd tangle link (Formula (1)):

$$A_n - A_{n-1} = 3^{n-1}\left(n \geq 1, n \in Z\right) \tag{1}$$

(ii) DNA Sierpinski links with even tangles

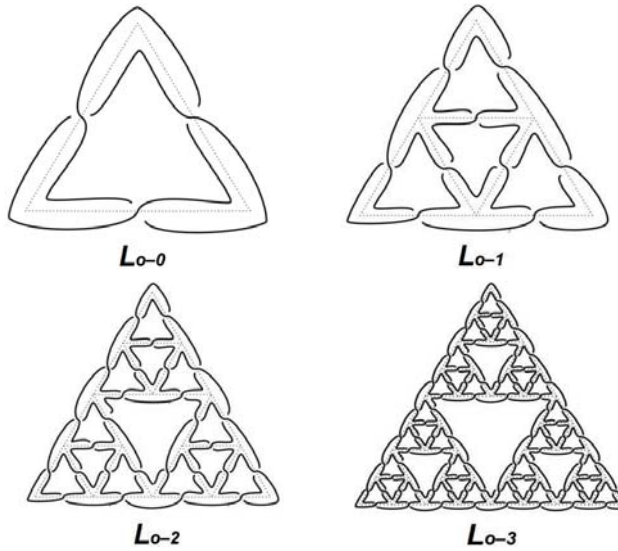

Figure 4. The first type DNA Sierpinski links with odd tangles, *n=0*, *1*, *2*, *3*.

Using even tangles to cover Sierpinski triangle E_n, it gets the link $L_{e\text{-}n}$. Figure 5 displays Sierpinski links covered by *k=2* tangles and the uncrossed vertexes in which *n* ranges from *0* to *4*. The results suggest a two components link rather than a trivial knot is obtained when *n* is zero. Similarly, there is an intrinsic law for the link growth when *n* increases. For the link $L_{e\text{-}0}$, it has 2 DNA strands, while $L_{e\text{-}1}$, $L_{e\text{-}2}$, and $L_{e\text{-}3}$ have 5, 14 and 21, respectively. Consequently, the growth of DNA Sierpinski links with even tangles can be expressed as:

$$A_n - A_{n-1} = 3^n (n \geq 1, n \in \mathrm{Z}) \tag{2}$$

(iii) DNA Sierpinski links with zero tangles

With respect to the Sierpinski triangles E_n covered by zero tangles, they are labeled as $L_{0\text{-}n}$. If *k* is zero, there are no crossings but parallel double lines. $L_{0\text{-}0}$, $L_{0\text{-}1}$, $L_{0\text{-}2}$, $L_{0\text{-}3}$ are shown in Figure 6. The component number A_0, A_1, A_2 and A_3 are 2, 5, 14 and 41, respectively. The growth mechanism of DNA Sierpinski links can be extrapolated which also satisfies Formula (2).

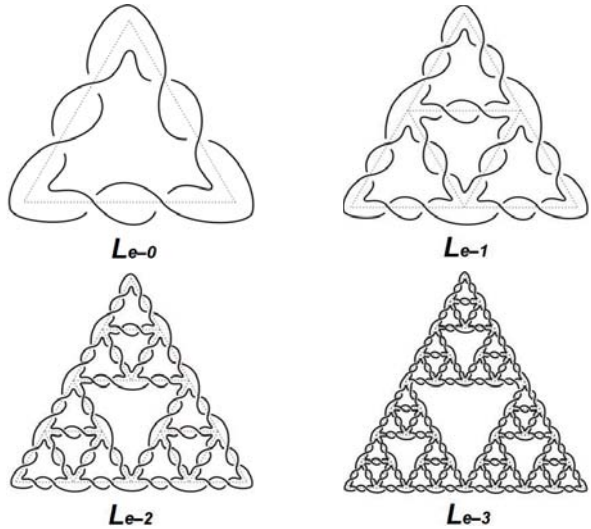

Figure 5. The first type DNA Sierpinski links with even tangles, *n=0*, *1*, *2*, *3*.

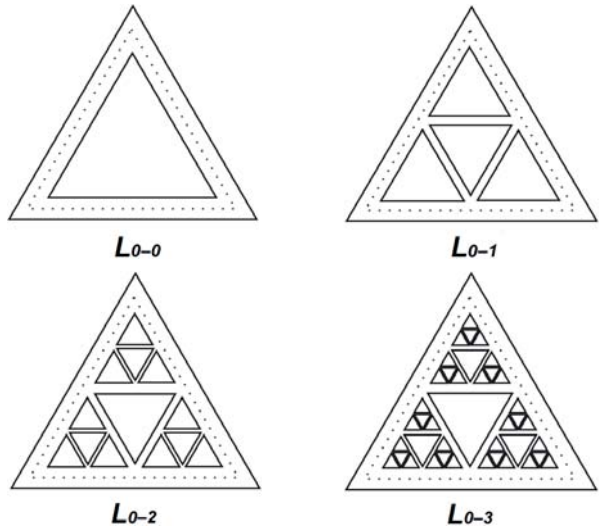

Figure 6. The first type DNA Sierpinski links with zero tangles, *n=0*, *1*, *2*, *3*.

It should be addressed here that the similar laws have been reported by Jiang and Jin [53]. The results obtained from our method are in good agreement with theirs, which confirms its validity.

2.2 The second type of DNA Sierpinski links

As stated above, the DNA Sierpinski links with uncrossed growing points are illustrated as follows. The *k=1, 2, 0* tangles are employed to discussed the cases DNA Sierpinski links with odd, even and zero tangles, respectively.

(i) DNA Sierpinski links with odd tangles

Different from the uncrossed growing points of the first type of DNA Sierpinski links, the growing points of the second ones are crossed, as depicted in Figure 7. Whatever the fractal number n is, the ultimate structure is a knot, meaning A_n always equals to 1. The interesting result implies the possibility of designing and synthesizing a series of DNA Sierpinski triangles with only one DNA strand.

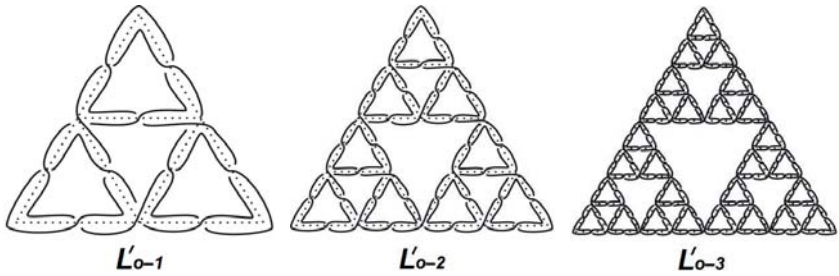

Figure 7. The second type DNA Sierpinski links with odd tangles, *n=1*, *2*, *3*.

(ii) DNA Sierpinski links with even tangles

As shown in Figure 8, a long scaffold DNA strand can be assembled into a framework where a certain amount of short DNA strands are nested, then a DNA Sierpinski triangle is gained. The component number A_n is found to be equal to 3^n+1. Because the number of the smallest triangles which with the same orientation of initial triangle E_0 is 3^n, A_n follows Formula (3).

$$A_n = 3^n + 1 (n \geq 1,\ n \in Z) \tag{3}$$

As a result, such fractal structures composed of a long DNA strand and some short DNA strands with same lengths may be designed and synthesized, and the first type of DNA Sierpinski links would be realized.

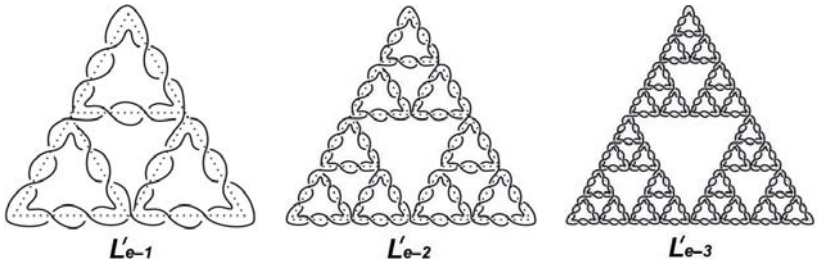

Figure 8. The second type DNA Sierpinski links with even tangles, *n=1*, *2*, *3*.

(iii) DNA Sierpinski links with zero tangles

Replacing uncrossed growing points of the first type DNA Sierpinski links with zero tangles with crossed ones, it can be found that A_n changes as the second type DNA Sierpinski link with even tangles does, as shown in Figure 9.

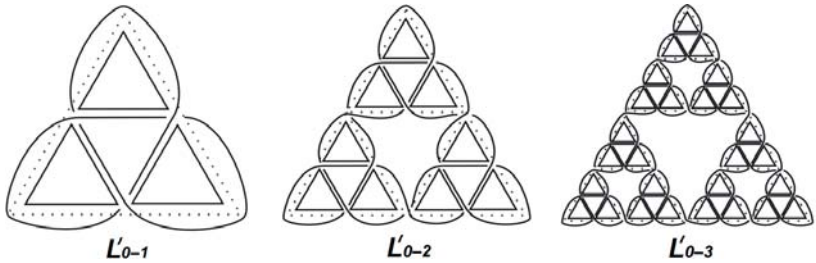

Figure 9. The second type DNA Sierpinski links with zero tangles, *n=1*, *2*, *3*.

3. Structure and properties

(i) Structural analysis of DNA Sierpinski links

To understand the properties of the DNA Sierpinski links, structural analysis is performed. For the first type DNA Sierpinski links with zero tangles, some different length DNA strands are designed, and then they are assembled to form the desired structures. If the growing points are crossed, a long scaffold DNA strand and some short DNA strands with same lengths appear. It is obvious that the numbers of self-intersect points are equal to those of growing points. We use C_n to indicate the number of self-intersect areas. It fulfills formula (4):

$$3C_{n-1}+3=3C_n \ (n\geq 2 \ \ and \ C_1=3) \tag{4}$$

The existence of self-intersect areas ensures that the structure is not unfastened. In principle, the more the self-intersect areas are, the steadier the structure is.

It is well-established that the twist number k changes with the fractal number n. It is a geometric sequence with common ratio *3* and the first term is *3k*. With regard to the first type DNA Sierpinski with odd tangles, there are three self-intersect areas in each DNA Sierpinski link, and they locate at the triangle areas close to the initial three vertexes (see Figure 4). If the growing points are replaced by crossed ones, k increases and equals to the number of the growing points. All points of the resulted knots are self-intersect areas that make DNA Sierpinski more stable. The twist number of the first type DNA Sierpinski links meets a geometric sequence with common ratio *3*, and the first term is *2k*. There is no self-intersect area in these structures. It means the stability arises from the interlocked DNA strands. The component number A_n will reduce $(3^n-1)/2$, and the self-intersect areas increases and equals to the number of growing points if the growing points are crossed. The stability of the structure is guaranteed by interlocked DNA strands and the self-intersect areas.

(ii) The designing rules of DNA strands

Herein, the concept of step length is defined. For an alternating links or knots, step length is the length from one over crossing (or under crossing) to the next over crossing (or under crossing). The full step length is the sum of step length of a DNA strand, the longest DNA strand is indicated by l_{max}.

Firstly, for the first type DNA Sierpinski links with odd tangles, if we suppose the length of each edge of Sierpinski is $0.5a$, then the step length of DNA strands are distributed as follows:

$n=0, 2a+2a+2a;$
$n=1, 2a+2a+2a;$
$2a+2a+2a+2a+2a+2a;$
$n=2, 2a+2a+2a;$
$2a+2a+2a+2a+2a+2a;$
$2a+2a+2a+2a+2a+2a+2a+2a+2a+2a+2a+2a;$
...

If n is arbitrary, the longest DNA strand is

$$l_{\max}=3\times 2^n\times 2a \quad (n\geq 0, n\in \mathrm{Z}) \tag{5}$$

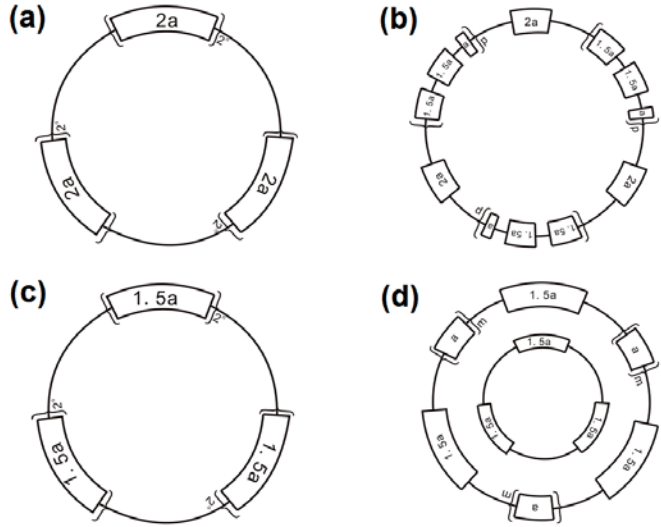

Figure 10. The models of DNA strands that are composed of DNA Sierpinski links.

The formula (5) suggests that any DNA Sierpinski links can be composed of DNA strands with l_{max} from longest to shortest, their numbers are 3^n, 3^{n-1}, 3^{n-2}, ..., *1* in order. Shown in Figure 10a is the corresponding DNA strand model. For example, the Sierpinski triangle with fractal number *2* is made of *6a*, *12a* and *24a* DNA strands, and their numbers are are 3^2, 3^1, 3^0, respectively.

If the uncrossed growing points are replaced by crossed ones, the distribution of DNA Sierpinski links is described below:

$$n=1, 2a+(1.5a+1.5a+a)+2a+(1.5a+1.5a+a)+2a+(1.5a+1.5a+a);$$
$$n=2, 2a+(1.5a+1.5a+a)_4+2a+(1.5a+1.5a+a)_4+2a+(1.5a+1.5a+a)_4;$$
...

It is clearly that $(1.5a+1.5a+a)_p$ is a repeated unit that always inserts between *2a* and *2a*. *p* satisfies formula (6), so the DNA strands model can be represented as shown Figure 10 b.

$$p=\frac{3^n-1}{2}\ (n\geq 0, n\in Z) \tag{6}$$

Secondly, with respect to the first type of DNA Sierpinski links with zero or even tangles, their distributions can be described below in terms of the above method:

$$n=0,\ \begin{cases}1.5a+1.5a+1.5a\\1.5a+1.5a+1.5a\end{cases};$$

$$n=1,\ \begin{cases}1.5a+1.5a+1.5a\\1.5a+1.5a+1.5a\\1.5a+1.5a+1.5a+1.5a+1.5a+1.5a\end{cases};$$

...

It shows that $(1.5a)_q$ is a repeated area and the longest DNA strand of any Sierpinski link is:

$$l_{\max}=3\times 2^n\times 1.5a\ \ (n\geq 0,\ n\in \mathrm{Z}) \tag{7}$$

This indicates that any DNA Sierpinski links can be made of DNA strands with l_{max} from longest to shortest and the number of them is 3^n, 3^{n-1}, 3^{n-2}, ... , 1 in order, the difference is that there are two styles of short DNA strands with equal full step length and their numbers are not same. For instance, the number of strands that constitute the smallest triangles with consistent orientation of initial triangle is 3^n, and the number of strands that constitute the smallest triangles with opposite orientation of initial triangle is 3^{n-1} (see Figure 6). Figure10c shows the DNA strands model, the closed circles indicate DNA strands, q satisfies formula (8):

$$q=3\times 2^n\ \ (n\geq 0,\ n\in \mathrm{Z}) \tag{8}$$

If the uncrossed growing points are taken placed by crossed ones, the distribution of DNA Sierpinski links is shown below:

$$n=1,\ \begin{cases}1.5a+1.5a+1.5a\\(a)_3+1.5a+(a)_3+1.5a+(a)_3+1.5a\end{cases};$$

$$n=2,\ \begin{cases}1.5a+1.5a+1.5a\\(a)_{12}+1.5a+(a)_{12}+1.5a+(a)_{12}+1.5a\end{cases};$$

$$n=3,\ \begin{cases}1.5a+1.5a+1.5a\\(a)_{39}+1.5a+(a)_{39}+1.5a+(a)_{39}+1.5a\end{cases};$$

...

The result shows that $(a)_m$ is a repeated area located between $1.5a$ and $1.5a$. The DNA strands model is shown in Figure 10 d, m satisfies formula (9):

$$3m_{n-1}+3=m_n\ \ (n\geq 1,\ n\in \mathrm{Z}) \tag{9}$$

(iii) The symmetry of DNA Sierpinski links

It is well known that the symmetry of Sierpinski triangles is C_{3v} and the step order is $2n$. Any DNA Sierpinski link will be superimposed with itself by rotating 120^0, indicating it has a three-fold symmetry axis. The DNA models shown in Figure 10 prove this result. Regardless of base-pairs, it can be concluded that each DNA strand possesses a three-fold symmetry axis

and the step order is n. It demonstrates that the symmetry of DNA Sierpinski link is reduced compared with Sierpinski triangle.

4. Investigation of synthesis method

A bottom-up synthesis method of DNA Sierpinski triangles is proposed in this paper based on the constructed structures.

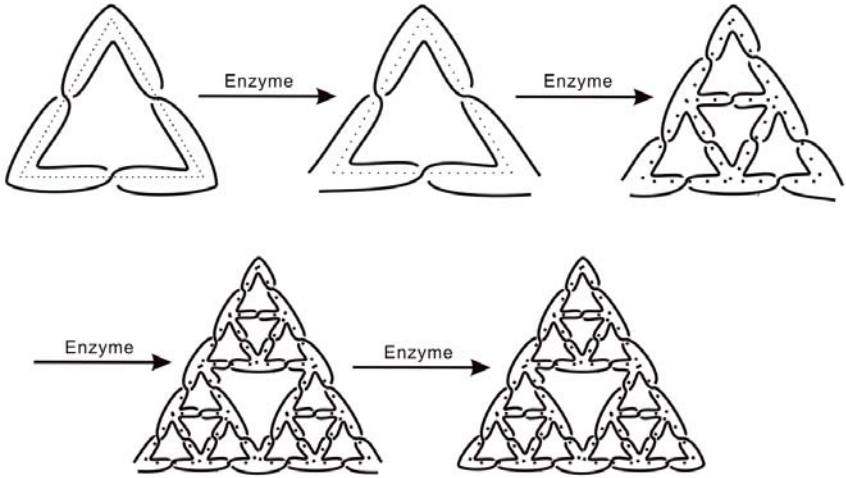

Figure 11. The strategy of synthesizing DNA Sierpinski links from basic building block.

Above all, the basic building block needs to be chosen. As shown in Figure 11, the $L_{o\text{-}0}$ on the leftmost is selected as the basic building block, the bigger and more complicate structures can be obtained by connecting some of them together. Second, special enzymes will perform well at the time. The chosen enzyme acts on two vertexes of the basic building block and breaks DNA strands. Then the sticky ends are bared outside and three basic building blocks are joined together by DNA Ligases, keep going and going, the last two cracks are also sewed up by suture enzyme then the desired structures will be produced. Any perfect DNA Sierpinski links will be realized by this novel method. In Figure 11, a DNA Sierpinski links with the fractal number 2 is synthesized from the basic building block $L_{o\text{-}0}$.

The strategy of synthesizing DNA Sierpinski links is useful for two dimensional structures; what about expanding it to three dimensional structures? The answer is yes but the basic building block is not $L_{o\text{-}0}$ anymore, a tetrahedron link is selected as basic building block. A schematic diagram is given in Figure 12, three tetrahedron links are connected together and

a 3D DNA Sierpinski links with fractal number 2 is shown as an example.

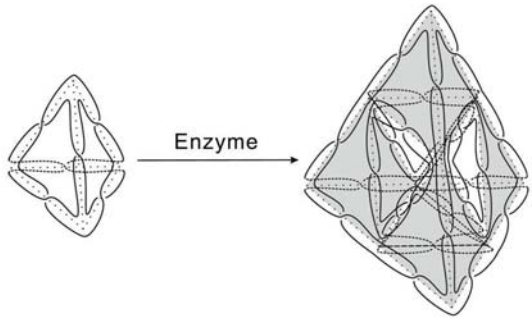

Figure 12. The strategy of synthesize 3D DNA Sierpinski links.

5. Conclusions

In this paper, a few tentative attempts were made on designing and characterizing DNA fractal links by mathematical methods. A series of DNA Sierpinski links were constructed by *k*-tangle models and two kinds of vertexes, and the growth mechanisms of two types of DNA Sierpinski links were unraveled by some formulas. We gave a definition of step length of DNA strands and analyze the distributions of step length, and then DNA strand models were got to describe the design rules of DNA strands.

A novel and exquisite bottom-up synthesis method based on the constructed DNA Sierpinski links with aid of enzymes was proposed. If we have gotten building blocks, then the desired structures can be realized with patience easily. The perfect method supplies a new train of thought for assembling two and three dimensional DNA fractal structures.

Although our study of DNA Sierpinski links and fractal links is only underway in theory, it gives an access for us to get a further understanding of macromolecular and viruses with fractal structures; these mathematic models maybe help chemists and biologists to checkout and develop their synthesis strategies.

Acknowledgements

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