# 4D printing method based on the composites with embedded continuous fibers

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#### Abstract

Most of the current 4D printing technologies have the following defects: 1) the deformation shape is simple; 2) the deforming precision is poor; 3) the deformation process is always uncontinuous. In this study, a new 4D printing process based on the composites with embedded continuous fibers is proposed. In this process, a bilayer structure consisting of the top layer of continuous fibers and the bottom layer with resin is 3D printed. Due to the different thermal expansion coefficient and elastic modulus of the top and bottom layers, the structure will produce bending deformation when the temperature changes. It is found that the curvature value and the curvature direction of the composite structure can be precisely controlled by the angle of the intersecting fibers. The influence of fiber trajectory on curvature is studied, and then, the controllable deformation of any developable surface is achieved.

**Keywords**: 4D printing, continuous fiber, composites, programmable morphing

## **1. Introduction**

4D printing is an additive manufacturing technology based on the development of 3D printing. 4D printed material can produce controllable deformation, and the factors that stimulate this deformation can be temperature, humidity and so on.<sup>[1]</sup> At present, there are three main methods for 4D printing. Shape memory materials are used as materials for 4D printing because they can change from temporary shapes to final shapes at the transition temperature.<sup>[2-4]</sup> However, this deformation is often discontinuous. Inspired by the deformation of pine cones and wheat awn, a new 4D printing method has been put forward.<sup>[5, 6]</sup> The rigid granular materials, such as cellulose fibers, are embedded in a flexible matrix, such as hydrogels. Because the rigid materials are inhomogeneous, the material will bend when it is placed in the water. However, this material can only achieve simple and imprecise deformation. If a variable external magnetic field or rheological field is applied during the printing process, a controllable orientation distribution of embedded particles can be obtained, and the complex shapes can be obtained.<sup>[7, 8]</sup> There are still several problems in this technology: first, the deformation accuracy of the material is not high; second, the magnetic field line can not be arbitrarily distributed; third, the printing process is complex.

In order to solve these problems, we developed the 4D printing technology based on continuous fiber embedded composites. The orientation of continuous fibers is easier to control, which will lead to higher deformation accuracy. In addition, this process is simpler and does not require complex outfield during the printing process. Through theoretical research and experimental verification, we have realized the deformation of complex developable surfaces.

# 2. Experimental section

#### 2.1 Materials and equipment

In this paper, the flexible matrix material used is Polyamide66 (PA66, SunDcreate Corp. in China), and the continuous fiber material is carbon fiber (Tangu Corp. in China). The 3D printer used in this experiment is used to print continuous fiber reinforced composites.<sup>[9]</sup> The fiber and resin can be fed into the heating head at the same time and fully impregnated. The composite material is directly extruded from the nozzle outlet. With this device, we can print out composite materials which can produce controllable deformation.

# 2.2 Preparation of continuous fibers embedded composites

The composite structure in this experiment is a bilayer. The bottom layer is resin, and the top layer is a mixture of continuous fibers and resin (shown in Figure 1). Because these two layers have different elastic moduli and coefficients of thermal expansion (CTEs), the composite structure can produce bending deformation when the temperature changes. As the specimen just removed from the substrate has internal stress, the heat treatment is necessary. The heat treatment temperature is 170-210 °C and the heat treatment time is 1-3 minutes.



Figure 1 4D printing process of continuous fibers embedded Composites

#### 2.3 Curvature measurement of surfaces

According to the experiment, when the fibers are distributed parallel on the surface of the composite, the material will be bent into a cylindrical shape. Therefore, we can measure the degree of deformation by measuring the curvature of the cylinder. When the radian of the cylinder is less than  $\pi$ , the equation to solve the curvature is:

$$kd/2 = \sin(kl/2) \tag{1}$$

where *d* is the chord length of the surface and *l* is the arc length of the surface. When the radian of the cylinder is more than  $\pi$ , a caliper can be

directly used to measure the diameter *d* and then to solve the curvature by:

$$k = \frac{2}{d} \tag{2}$$

# 3. Results and discussion

# 3.1 Deformation mechanism of the composite material

When the fiber bundles are distributed parallel on the resin surface, the composite structure will cause bending deformation due to the different CTEs and elastic moduli of the continuous fibers and resin, as shown in Figure 2.



Figure 2 The composite with parallel continuous fibers embedded

Through the modulus of the resin is negligible compared to the modulus of the continuous fibers, the strain energy on the surface is approximately equal to the sum of the strain energies of the two columns of fiber bundles:

$$\Delta e \approx \frac{1}{2} E(\Delta \varepsilon_a^2 + \Delta \varepsilon_b^2) \tag{3}$$

where  $\Delta \varepsilon$  is the strain energy difference between when two columns of fiber bundles exist together and when one column of fiber bundles exist alone.

For a point in the composite structure, assuming that its distance from the neutral surface is d, and the length of the neutral surface is  $l_0$ , therefore:

$$\Delta \varepsilon = \varepsilon' - \varepsilon_0 = d(k' - k_0)l_0 \tag{4}$$

where  $k_0$  and  $\varepsilon_0$  are the curvature of the neutral surface and the strain at the height of that point when a column of fiber bundles exists alone. k'and  $\varepsilon'$  are the curvature of the neutral surface and the strain at the height of that point when two columns of fiber bundles exist together. Therefore,

$$\Delta e \propto (k_a - k_{a0})^2 + (k_b - k_{b0})^2 \tag{5}$$

According to the Euler formula of differential geometry, the following equation can be obtained:

$$\Delta e \propto (k_a - k_1 \cos^2 \alpha)^2 + (k_b - k_1 \cos^2(\theta - \alpha))^2 \tag{6}$$

where  $\theta$  is the angle between two fiber bundles and  $\alpha$  is the angle between the direction of the principal curvature and one of the fiber bundles.

For our 4D printing process,  $k_a$  and  $k_b$  are all the curvature when a column of fiber bundles exists alone, so  $k_a=k_b$ . In order to make the  $\Delta e$  minimum, the  $\alpha$  and  $k_1$  (the principal curvature of the surface) can be solved:

$$\begin{cases} \alpha = \frac{\theta}{2} \\ k_1 = \frac{k_a}{\cos^2 \frac{\theta}{2}} \end{cases}$$
(7)

To verify this equation, we printed rectangular composite panels with different angles ( $30^\circ$ ,  $60^\circ$  and  $90^\circ$ ) between two columns of fiber bundles.



**Figure 3** The direction of the curvature is along the angular bisector between fibers The relationship between the size and the direction of the principal

curvature and the angle of fiber bundles are shown in Figure 4 and 5.



Figure 4 The relationship between the size of the curvature and the angle of fiber bundles



Figure 5 The relationship between the direction of the curvature and the angle of fiber bundles

According to the theory and experiment, we can get the law that the principal curvature direction of the bilayer surface is in the direction of the bisector of the sharp angle between the fiber bundles, and the principal curvature value of the bilayer surface is proportional to  $\sec^2\theta$ . With this rule, we can design more complex developable surfaces.

#### 3.2 Deformation design of a complex surface

Let's take a conical surface  $r = (u \cos v, u \sin v, u \cot \gamma)$  as an example, where *u*, *v* are two parameters of the cone, and  $\gamma$  is the half vertex angle of the cone. Solve the value and the direction of the non-zero principal curvature, and the value of the non-zero principal curvature is:

$$k_1 = \frac{\cos \gamma}{u} \tag{8}$$

It can be seen that the curvature of the position closer to the conical vertex is larger, as shown in Figure 6a. The direction of the curvature is along the direction of the weft of the cone (Figure 6b).



**Figure 6** The size and the direction of the curvature of a cone The cone is changed into a plane by isometric transformation, and

the principal curvature can be expressed by the plane coordinate system  $(\rho, \theta)$ , as shown in Figure 7.



Figure 7 The principal curvature line after isometric transformation

Rearrange Equation 7 and 9, then the angle  $\lambda$  between the orientation of fibers and the direction of non-zero principal curvature can be obtained:

$$\cos \lambda = \pm \sqrt{\frac{k_a}{k_1}} = \pm \sqrt{\frac{\rho k_a}{\cot \gamma}}$$
(10)

In polar coordinates,

$$\tan \lambda = \frac{\rho'}{\rho} \tag{11}$$

Rearrange Equation 10 and 11, then the equation for the fiber trajectory can be obtained:

$$\frac{\mathrm{d}\rho}{\mathrm{d}\theta} = \pm \sqrt{\rho(\frac{\cot\gamma}{k_a} - \rho)} \tag{12}$$

Solve the differential equation, therefore:

$$\rho = \frac{1}{2k_a \cot \gamma} [1 + \sin(\pm \theta + C)] \tag{13}$$

where *C* is an arbitrary constant that determines the position of the curve, and the obtained fiber trajectories are shown in Figure 8.



Figure 8 The obtained fiber trajectories

In order to verify this equation, we take the appropriate C values, so that the distance between adjacent family of lines is near 8 mm. Print the composite structure according to the designed carbon fiber trajectory and heat the material to get the deformed structures, as shown in Figure 9.



Figure 9 The cone prepared by 4D printing

In order to verify the deformation accuracy, we printed many conical surfaces with designed fiber trajectories and cylindrical surfaces with parallel fiber trajectories. In each group of experiments, the printing parameters and the heat treatment parameters of the cone and the cylinder are exactly the same. The  $K_a$  calculated by the curvature of the bottom surface of a conical surface is called the calculated curvature, and the curvature measured by a cylindrical surface is called the experimental curvature. The two curvatures are compared, and the results are shown in Table 1. It can be seen that the error between calculated curvature and experimental curvature is between 3-8 %, indicating higher deformation accuracy.

Table 1 Comparison of experimental curvature and calculated curvature

Group	Experimental curvature	Calculated curvature	Error
1	0.02477	0.02295	7.35%

2	0.01511	0.01457	3.57%
3	0.06670	0.06119	8.26%
4	0.04833	0.04543	6.00%

According to this method, any developable surface can be prepared by this 4D printing process. The specific solution is still to get the main curvature lines of the curved surface first, then transform it into a plane curve by isometric transformation, and finally get the fiber trajectory.

## 4. Conclusion

We used 4D printing technology to prepare continuous fiber embedded composites. Due to the different elastic moduli and CTEs of continuous fibers and resin, the composite structure will have different flexural curvature at different temperatures. It is found that when the fiber bundles are aligned in parallel, the direction of principal curvature is along the fiber orientation. When two columns of fiber bundles are distributed on the surface of the resin, the direction of curvature is along the bisector of the angle  $\theta$  between the intersecting fiber bundles, and the size of the curvature is proportional to sec<sup>2</sup> $\theta$ . With this rule, we can design more complex surfaces, such as a cone. This 4D printing process has good designability and deformation accuracy.

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