Bending Behavior of Shape Memory Polymer Based Laminates

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ABSTRACT

Shape memory polymers (SMP) are smart materials was characterized by the recoverability of shape memory effect, but its mechanical property such as the strength is low. In this study, for industrial applications, a carbon fiber fabric reinforced shape memory polymer was developed. Four kinds of specimens with different laminations of carbon fiber fabric and shape memory polymer sheet were prepared. The bending recoverability was investigated and compared between the SMP sheet substance and the developed SMP based laminates. Both of the materials were loaded and then unloaded repeatedly above the Tg. The bending recoverability characterized by the bending angle, and the influence of weight of specimen was examined. Results show that the SMP based laminates developed have good shape recoverability, and their bending recovery ratio was larger than that of the SMP sheet at any recovery time. Furthermore, the bending recovery ratio was predicted with an analysis model and its result was compared with experiment values. The good agreement between the experiment and analysis suggests that the proposed analysis model is effective for the prediction of the bending recovery of SMP based laminates.

Keyword: Shape memory polymer, Composites, Carbon fiber fabric, Bending recovery

1. INTRODUCTION

Shape memory material has the property to return mechanically induced strains upon heating or application of an electric field. The typical shape memory materials are shape memory alloys and shape memory polymers (SMP). SMP's have the characteristics such as large recoverability, lightweight, superior molding property and lower cost. These advantages have resulted in the SMP's becoming functional materials which have attracted attention from many fields. For most of the SMP's, their glass transition temperature (Tg) may be set up around the room temperature, and the characterizations such as shape recovery and/or shape fixation may appear at the temperatures above and/or below Tg [1-9]. Figure 1 shows a shape memory process of SMP material. Its process may consist of following steps [1-3,9,10]:

1. Form the SMP at fluxing temperature and cool down it below Tg.
2. Deform the formed shape above Tg.
3. Fix the deformed shape and cool it below Tg and then remove the constrain from SMP
4. Heat SMP above Tg to recover original shape.

Among various SMP's, a shape memory polymer of polyurethane series is one of the most popular shape memory polymers since its glass transition temperature can be set up at any temperature within ±50K around room temperature. It has a large difference in mechanical properties, optical characteristics and steam permeability above and below the temperature Tg. Thus an SMP of polyurethane series will have wide applications in many fields of industry, medical treatment, welfare and daily life [11-17]. Although SMP materials already have some applications in the engineering industry, they are far from the technological potential of SMP. The main reason for this is the significant weaknesses in mechanical strength and stiffness compared with metals and ceramics. On the other hand, adding some reinforcements to the SMP matrix allows easy tailoring of the material stiffness. A few researchers have studied composite materials based on a shape memory polymer matrix [5-7,18]. These studies demonstrated that glass fiber reinforcements increased the stiffness of the SMP resins and reduced recoverable strain levels.

In this paper, for the wider use of the SMP's, the composites based on SMP films and carbon fiber fabric were developed. The sequence and position of the SMP film and carbon fiber fabric in a laminate were changed, and the optimization of laminated structure was discussed. The dynamic mechanical analysis (DMA) was conducted and shape recoverability and shape fixation characterization were investigated. The original shape of SMP based laminates was recovered by changing the temperature above and/or below Tg. The bending recoverability for both the SMP sheet and the developed composites under different loading and heating conditions was examined. Moreover, the bending recovery ratio was predicted and its result was compared with experiment values. The effectiveness of analysis model is discussed.

2. EXPERIMENTAL WORK

2.1 Raw Materials and Fabrication of Shape Memory Polymer Based Laminates

The polyesterpolyol series of polyurethane SMP (Diary, MS4510) was used and its glass transition temperature Tg was about 45˚C. The raw material was liquid. The curing agent was dimethylformamide (DMF). The weight ratio of polymer to DMF was set to be 3:7.

The SMP films were molded by the dry film forming method as shown in Fig.2. The procedure is as follows. Polyester film was spread on an iron plate, and the spacer with specified thickness was put in both sides. The solution was poured and enlarged with the brass stick at a constant force and speed. The cure conditions were 70˚C 1 hour, 100˚C 1 hour and 110˚C 2 hours. After solvent volatilized from the solution and SMP film was obtained. For the fabrication of CF fabric prepreg, the woven carbon fiber fabric (Toray Industries, Inc., CO 6343) was laid on polyester film, SMP solution was poured, and the CF fabric prepreg with carbon fiber fabric was obtained. Then, the SMP films and CF fabric prepreg were laminated so that specimens of both SMP sheet and SMP based laminates were fabricated. The hot press process was conducted in the 7 ton compression machine under the vacuum bag and molding compression pressure of 1.3MPa. Four kinds of specimens were developed as shown in Fig.3.

2.2 Experimental Method

2.2.1. Dynamic mechanical analysis (DMA)

In order to examine the thermal behavior of SMP based laminates, the dynamic mechanical rheological test was conducted. Dynamic mechanical rheological measurement equipment (Rheogel-E4000) was used. The DMA behavior of the SMP sheet and SMP based laminate (SMP/CF fabric/SMP) was measured. The samples dimension is 30×5×2.5 mm. In the DMA measurement, the dynamic tensile mode with a frequency of 1Hz was performed at a fixed stain of 0.1%. Temperature rang was set from 0˚C to 100˚C using a heating rate of 2˚C/min.

2.2.2. Bending recovery test

Before doing bending recovery tests, the specimen was bent at a specified angle with the jig as shown in Fig.4. The samples with dimensions of 80×10×2.5 mm were taken from the fabricated laminates. One end of the specimen was fixed with 20 mm length on the jig and the other end was deformed with the length of 60 mm. Then, the whole jig with the specimen fixed on it was put into the temperature-controlled chamber at 65˚C for 1 hour. The specimen was covered along the jig shape by the polyester film and then was cooled down to 15˚C. Then, the specimen with a fixed bending angle was produced due to the shape fixation of SMP.

Recovery bending test were accomplished using temperature-controlled chamber. One end of the specimen was hold with 20 mm length and then the specimens were heated from 15˚C to 65˚C at the heating rate of 10˚C /min. The recoverability was measured and the recovering curve was recorded.

The time dependency of bending recovery property was investigated under the both recovery status with and without the self-weight. The specimen was kept at 65˚C for 5 minutes, 10 minutes, 15 minutes and 20 minutes, respectively. In order to investigate the influence of self-weight, four initial bending angles of -45˚, 0˚, 45˚ and 90˚ were designed and the specimens with these initial angles were set up at the temperature of 65˚C for 20 minutes.

3. RESULTS AND DISCUSSIONS

3.1 Dynamic Mechanical Property for SMP Based laminates

The DMA property of SMP based laminates with carbon fiber fabric and SMP sheet is shown in Fig.5. It is clear that there existed the temperature dependency of storage modulus

E', loss modulus E'' and loss factor tan δ for these two material. The loss modulus E'' was smaller than storage modulus E' for both material. Glass transition temperature region of SMP sheet was 20˚C~55˚C, while it is 25˚C~60˚C for SMP/CF fabric/SMP material. These showed that Tg moved to the higher temperature side. The onset of tan δ curve appears around 35˚C. The peak tan δ of SMP based laminate is little lower than that of SMP sheet. The maximum of E'' for both materials existed and its peak value for both modulus curve employed to define the glass transition temperature Tg. Tg of SMP was about 39˚C while SMP/CF fabric/SMP material was 46.3˚C from loss modulus E'' curve.

A large change above and below Tg for storage modulus E' was observed for about 100 times. The key point for shape memory effects of SMPs was how to utilize and control the variation of E' at the temperature below and above Tg. This indicates that SMP transforms easily above Tg, while the resistance for deformation is large below Tg. This also showed that SMP had shape fixation property and shape recovery property. The developed composite material has large storage modulus E' compared with a SMP sheet. Especially, at the temperature about Tg, storage modulus E' was much larger than that of SMP sheet. It is clear that the elastic modulus of SMP based laminates was increased since polyurethane SMPs were reinforced by carbon fiber fabric.

3.2 Bending Recovery Property for SMP Based Laminates

In order to examine the bending recovery ability of SMP based laminates, the bending recovery ratio was defined by the parameter, $R = \omega_R / \omega_0$, where ω_R was defined as the recovery deflection and ω_0 was the initial deflection as shown in Fig.6.

It should be noted that the test temperature of 15˚C was below glass transition temperature region of the material, while the temperature of 65˚C was above glass transition temperature region.

3.2.1 Bending Recovery Property with Different Recovery Time

For both recovery statuses with or without the self-weight in SMP sheet and SMP based laminates, the time dependency of the bending recovery property was investigated. The relationship between the bending recovery ratios and recovery time with or without the self-weight was shown in Fig.7 and Fig.8, respectively. Their recovering process photographs are shown in Fig.9 and Fig.10, respectively.

From Fig. 7 to Fig.10, it is clear that the SMP based laminates developed have good shape recoverability. The recovery ratio of the SMP/CF fabric specimen was larger than other specimen at the same recovery time. Since the elastic stiffness of the carbon fiber fabric and SMP differ, the stress of each layer varies according to lamination theory. Then a bending moment will be produced for an unsymmetrical lamination. Since the direction of resulted bending moment is the recovery direction of SMP/CF fabric specimen, the rate of recovery becomes overshot and the value of *R* in Fig. 7 became larger than 100%. Oppositely, the rate of recovery for CF fabric/SMP is small. As for the influence of the recovery time, the recovery ratio increased with increments of recovery time, but after 15 minutes became smaller and smaller.

For the cases with or without self-weight, the specimens were set up in vertical positions as shown in Fig. 9 to avoid the influence of self-weight, while the specimens were set up in horizontal status in Fig. 10 to take into account the influence of self-weight. The recovery rate in Fig. 9 without the influence of self-weight was larger than that in Fig. 10. It is clear that a great influence is exerted on bending recovery due to self-weight of specimen.

3.2.2 Influence of Initial Angle on Bending Recovery Property

The relationship between the bending recovery ratio and the initial angle was shown in Fig.11 and the recovering status with different initial angles of four specimens was shown in Fig.12.

From these figures, it is found that the recovery ratio was the lowest at the initial angle 0˚, while it is the largest at the initial angle 90˚ for SMP sheet, SMP/CF fabric/SMP and CF fabric/SMP. The recovery ratio is almost same for both initial angle of 45˚ and -45˚. These results indicated the great influence of the initial angle on the recoverability. However, the recovery ratio for SMP/CF fabric is over 100% and it is definitely higher than other materials. However. The recovery ratio is almost same at any initial angle because in this case the recovery force was relatively small to bending moment.

4. ANALYSIS OF BENDING RECOVERY RATIO

4.1 The bending stiffness of CF fabric prepreg

For explication of the stiffness of the laminates, bending stiffness $E_{CF}I_{CF}$ of CF fabric prepreg was calculated. For CF fabric prepreg, it is like a cantilever beam and its bending stiffness $E_{CF}I_{CF}$ can be calculated with the reference of Fig.13. When the distance s of the specimen is measured from origin point O, the curvature is represented by $d\theta/ds$. Differential equation of bending curve is

$$E_{CF}I_{CF}\frac{d\theta}{ds} = -Wx \qquad (1)$$

If this equation is differentiated to s, where the abscissa axis x and distance s is related to the following equation

$$\frac{dx}{ds} = \cos\theta$$

Therefore equation (1) becomes

$$E_{CF}I_{CF}\frac{d^2\theta}{ds^2} = -W\cos\theta \qquad (2)$$

$d\theta$ is multiplied on both sides, and equation (2) is integrated, so that

$$\int \frac{d^2\theta}{ds^2} \bullet \frac{d\theta}{ds} ds = -\frac{W}{E_{CF} I_{CF}} \int \cos\theta d\theta$$

This can also be written by

$$\frac{1}{2}\int \frac{d}{ds}\left(\frac{d\theta}{ds}\right)^2 ds = -\frac{W}{E_{CF} I_{CF}} \int \cos\theta d\theta$$

If this equation is integrated, then

$$\frac{1}{2}\left(\frac{d\theta}{ds}\right)^2 = -\left(\frac{W}{E_{CF} I_{CF}}\right)\sin\theta + C$$

Since bending moment is θ at a free end, $d\theta/ds = 0$ and $\theta = \alpha$, so that

$$C = \left(\frac{W}{E_{CF} I_{CF}}\right)\sin\alpha$$

Therefore,

$$\left(\frac{d\theta}{ds}\right)^2 = 2\left(\frac{W}{E_{CF} I_{CF}}\right)(\sin\alpha - \sin\theta)$$

that is,

$$\frac{d\theta}{ds} = \pm\sqrt{2}\sqrt{\frac{W}{E_{CF} I_{CF}}(\sin\alpha - \sin\theta)}$$

From Fig.13 since $d\theta/ds$ is always negative, it removes a positive from this equation,

then

$$ds = -\frac{d\theta}{\sqrt{2}\sqrt{\frac{W}{E_{CF} I_{CF}}(\sin\alpha - \sin\theta)}}$$

If the limit of integration is exchanged, the full length of specimen is

$$l = \int ds = \int_0^{\alpha} \frac{d\theta}{\sqrt{2}\sqrt{\frac{W}{E_{CF} I_{CF}}(\sin\theta - \sin\alpha)}}$$

this is

$$E_{CF}I_{CF}=\left(\frac{l}{\int_0^{\alpha}\frac{d\theta}{\sqrt{2}\sqrt{W(\sin\alpha-\sin\theta)}}}\right)^2 \quad (3)$$

where $l=0.06m$, $W=0.02N$ in the practical test. α is measured by experiment at 65°C and 15°C. α_H measured at 65°C is 29.32° and α_L at 15°C is 16.4°. The bending stiffness $E_{CF}I_{CF}$ then is calculated at 65°C and 15°C, respectively, and its values are $(E_{CF}I_{CF})_H=6.37\times10^{-5}N\cdot m^2$ and $(E_{CF}I_{CF})_L=1.22\times10^{-4}N\cdot m^2$, respectively.

4.2 Recovery moment of SMP based laminates

Under the recovery status without the influence of self-weight, the recovery moment of SMP base laminates is appeared by the following equation.

$$M=M_1+M_2 \quad (4)$$

where M_1 is the recovery moment of SMP and M_2 is the moment resulted from the variation of shear stress at different laminas

The relationship between the recovery moment of SMP and curvature radius can be related by the following equation.

$$\begin{aligned}M_1&=E_{SL}I_S/R_{SL}-E_{SH}I_S/R_{SH}\\&\approx E_{SL}I_S/R_{SL}\end{aligned} \quad (5)$$

where R_{SL} and R_{SH} are curvature radius at 15°C and 65°C, respectively. E_{SL} and E_{SH}

are Young's modulus at 15°C and 65°C. I_S is second moment of cross-section.

On the other hand, the cross-section of laminates is shown in Fig.14. The moment within a SMP based laminate at 65° can be expressed according the laminated theory [20]

$$M_2 = D_x k \tag{6}$$

where D_x is bending stiffness matrix and k is curvature of middle surface.

In equation (6) D_x is given by

$$\begin{aligned} D_x &= \frac{1}{3}\sum_{k=1}^{2} Q_x^{(k)}(z_k^3 - z_{k-1}^3) \\ &= \frac{1}{3}\left[Q_x^{[1]}(z_1^3 - z_0^3) + Q_x^{[2]}(z_2^3 - z_1^3)\right] \end{aligned} \tag{7}$$

The elastic modulus $Q_x^{[1]}$ and $Q_x^{[2]}$ are defined as

$$Q_x^{[1]} = \frac{E_{CF}}{1-\nu_{CF}^{\ 2}}$$

$$Q_x^{[2]} = \frac{E_{SH}}{1-\nu_S^{\ 2}}$$

where ν_{CF} is Poisson's ratio of CF fabric prepreg, and ν_{CF} is assumed to be 0 due to fabric, and ν_S is Poisson's ratio of SMP resin. Therefore, equation (7) becomes

$$\begin{aligned} D_x &= \frac{1}{3}\left[\left(1-\nu_S^2\right)^{-1} E_{SH} \bullet \frac{h^3+3ht^2}{4} + E_{CF} t \bullet \frac{3h^2+t^2}{4}\right] \\ &= \frac{1}{3}\left[\left(1-\nu_S^2\right)^{-1} E_{SH} \bullet \frac{h^3+3ht^2}{4} + E_{CF} I_{CF} \bullet \frac{12}{bt^2} \bullet \frac{3h^2+t^2}{4}\right] \\ &= \frac{1}{3}\left[\left(1-\nu_S^2\right)^{-1} E_{SH} \bullet \frac{h^3+3ht^2}{4} + E_{CF} I_{CF} \bullet \frac{3}{bt^2} \bullet (3h^2+t^2)\right] \end{aligned} \tag{8}$$

Then, the curvature of middle surface is obtained by the following equation.

$$k = -\frac{\partial^2 \omega_0}{\partial x^2} \qquad (9)$$

where ω_0 curve is measured in experiments and so k is obtained.

4.3 Bending recovery ratio of SMP based laminates

The equivalent cross section for SMP based laminates is calculated with the property of SMP sheet. The shape of SMP based laminates was recovered by changing the temperature (see Fig.6). If the second moment of equivalent cross section is marked by I_{eL} at low temperature and I_{eH} at high temperature, the relation between recovery moment and change of curvature for whole SMP based laminates can be written by

$$M = E_{SL} I_{eL} / R_L - E_{SH} I_{eH} / R_H \qquad (10)$$

The recovery moment *M* can be obtained from the equation (4) according to the above analysis. The equivalent cross section of the specimen is shown in Fig.15. The width d of CF fabric is equivalent to be b_L with SMP sheet at low temperature. So, b_L is given by

$$b_L = \frac{E_{CF}}{E_{SL}} b = \frac{1}{E_{SL}} \bullet \frac{12 E_{CF} I_{CF}}{t^3}$$

In Fig. 15, the equivalent cross section of the SMP based laminate specimen can be divided into two rectangles, and according to the area moment approach the following equation is effective, where h_{cL} is the distance from centroid *C*.

$$(b_L \times t + b \times h) \times h_{cL} = (b_L \times t) \times \left(\frac{t}{2} + h\right) + (b \times h) \times \frac{h}{2}$$

Thus, the second moment of equivalent cross section I_{cL} at low temperature can be written by

$$I_{eL}=\left[\frac{bh^3}{12}+bh\left(h_{cL}-\frac{h}{2}\right)^2\right]+\left[\frac{b_L t^3}{12}+b_L t\left(h-h_{cL}+\frac{t}{2}\right)^2\right]$$

And the second moment of equivalent cross section I_{eH} at high temperature can be represented by

$$I_{eH}=\left[\frac{bh^3}{12}+bh\left(h_{cH}-\frac{h}{2}\right)^2\right]+\left[\frac{b_H t^3}{12}+b_H t\left(h-h_{cH}+\frac{t}{2}\right)^2\right]$$

From equation (10), this can be rewritten as follows.

$$R_H=\frac{E_{Sh}I_{eH}}{E_{Sl}I_{eL}/R_L-M}$$

$$\omega_R=\omega_0-R_H\left[1-\cos\left(\frac{l}{R_H}\times\frac{180}{\pi}\right)\right]$$

$$R=\frac{\omega_R}{\omega_0}\times 100\% \qquad (11)$$

Thus, according to the above process, the bending recovery ratio of SMP base laminates can be predicted.

4.4 Comparison of bending recovery ratio between analysis and experiments

The geometrical and mechanical parameters of SMP sheet are shown in Table 1. The bending recovery behavior of SMP based laminates was measured by changing the test temperature below and above Tg. Both the calculation and experiment results of bending recovery ratio are listed in Table 2. It is indicated that the analysis prediction gives a reasonable result for each material.

5. CONCLUSIONS

In the study, shape memory polymer (SMP) based laminates with carbon fiber fabric were developed, and their bending recovery property was examined by changing recovery time and initial angles. The results obtained are remarked as follows.

(1) The storage modulus E' has a large change above and below Tg. Its value in SMP based laminates was much higher than SMP sheet.

(2) The developed SMP based laminates have excellent shape recoverability.

(3) The bending recovery ratio of the SMP based laminates was larger than that of SMP sheet at each recovery time. Recovery time has a great influence on the bending recoverability. The bending recovery ratio became large with the increment of recovery time.

(4) The bending recovery ratio will be the smallest at initial angle 0˚ and the largest at 90˚ for SMP sheet, SMP/CF fabric/SMP and CF fabric/SMP composites. It is shown that the weight of specimen has a significant influence on bending recovery ratio.

(5) The analysis model for the prediction of bending moments in the materials developed shows that a reasonable result is obtained when compared with the experimental evidence.

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laminates

Fig.12. The bending recovery status for different specimens under various initial angles

Fig.13. Bending stiffness $E_{CF}I_{CF}$ for CF fabric prepreg

Fig.14. Cross section for SMP based laminates

Fig.15. Equivalent cross section of SMP based laminates

Table Ⅰ Geometrical and mechanical parameters

Table Ⅱ Bending recovery ratio for SMP base laminates

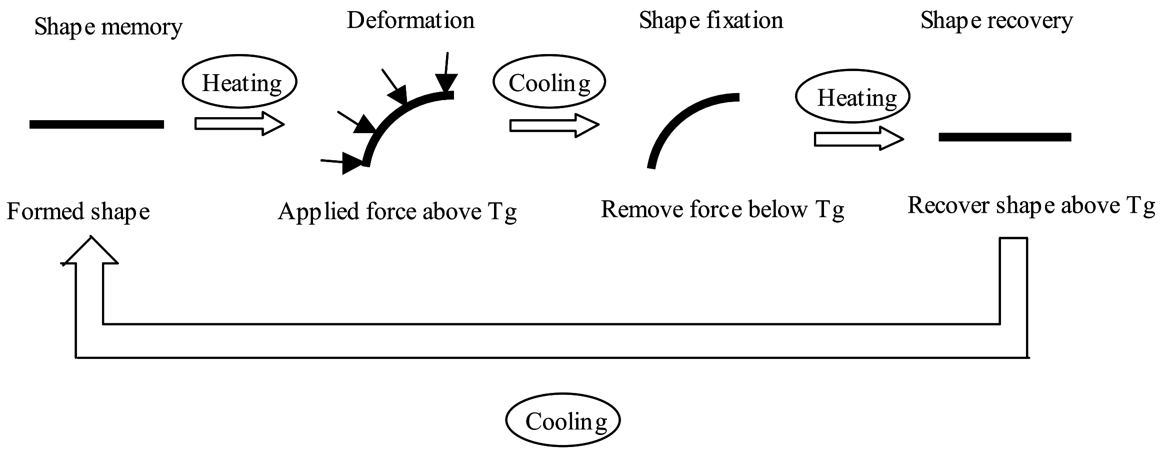


Fig.1. Shape memory process of a SMP material

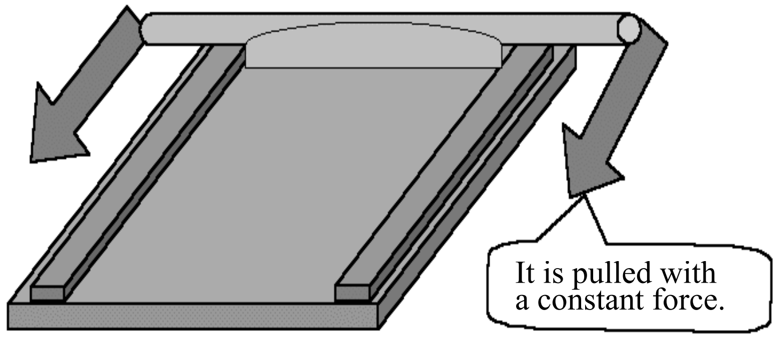


Fig.2. Schematic illustration for film forming

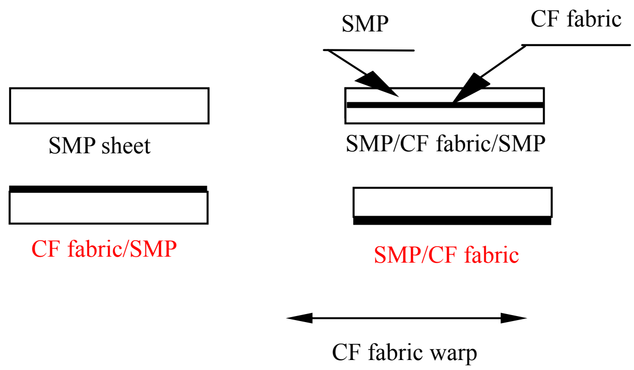


Fig.3. Types of specimens

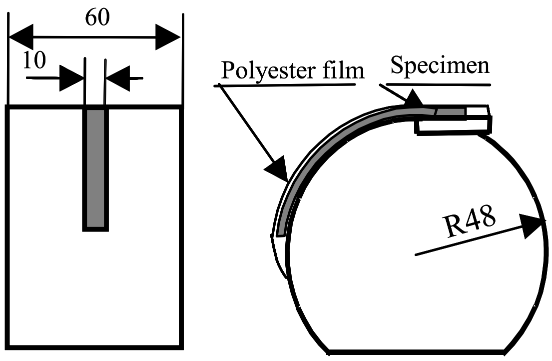
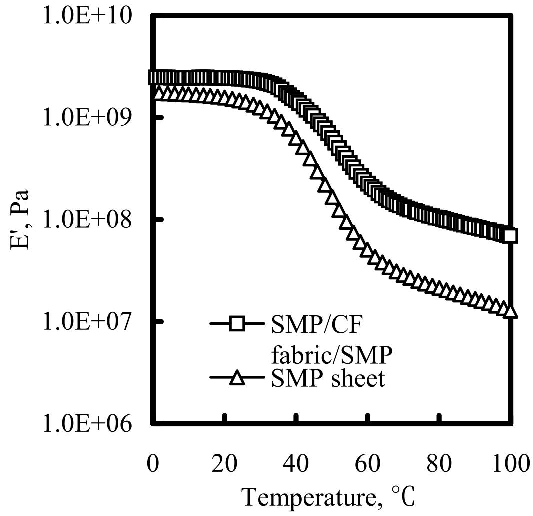
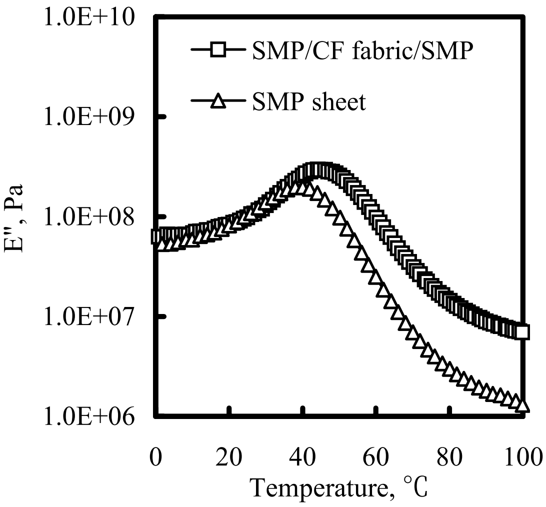


Fig.4. The jig for fixing specimen shape



(a)



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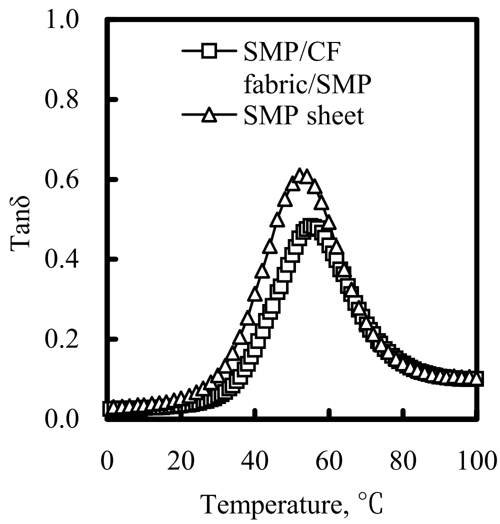


(c)

Fg.5. Dynamic mechanical properties: (a) Storage modulus E'
(b) Loss modulus E" (c) Loss factor tan δ

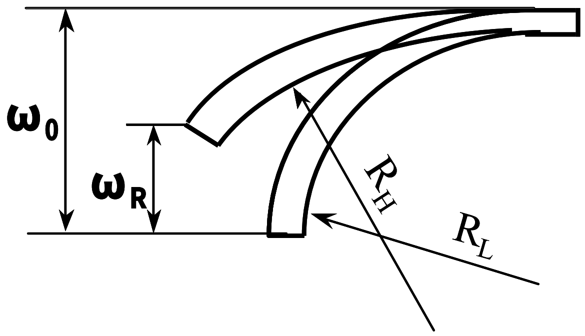


Fig.6. The definition of bending recovery ratio

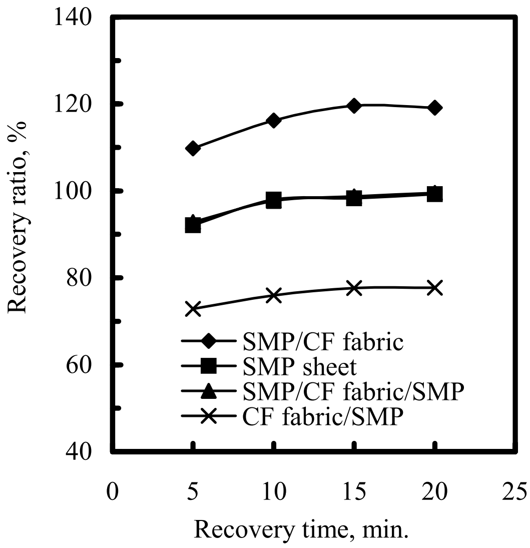


Fig.7. The relationship between the recovery ratios and recovery time under the status without the self-weight of specimen

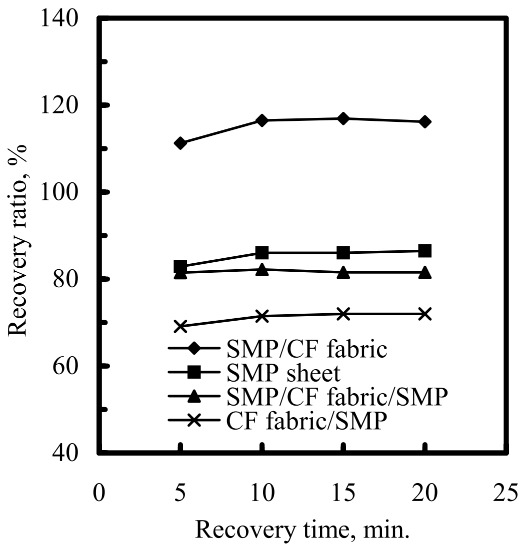


Fig.8. The relationship between the recovery ratios and recovery time with the self-weight of specimen

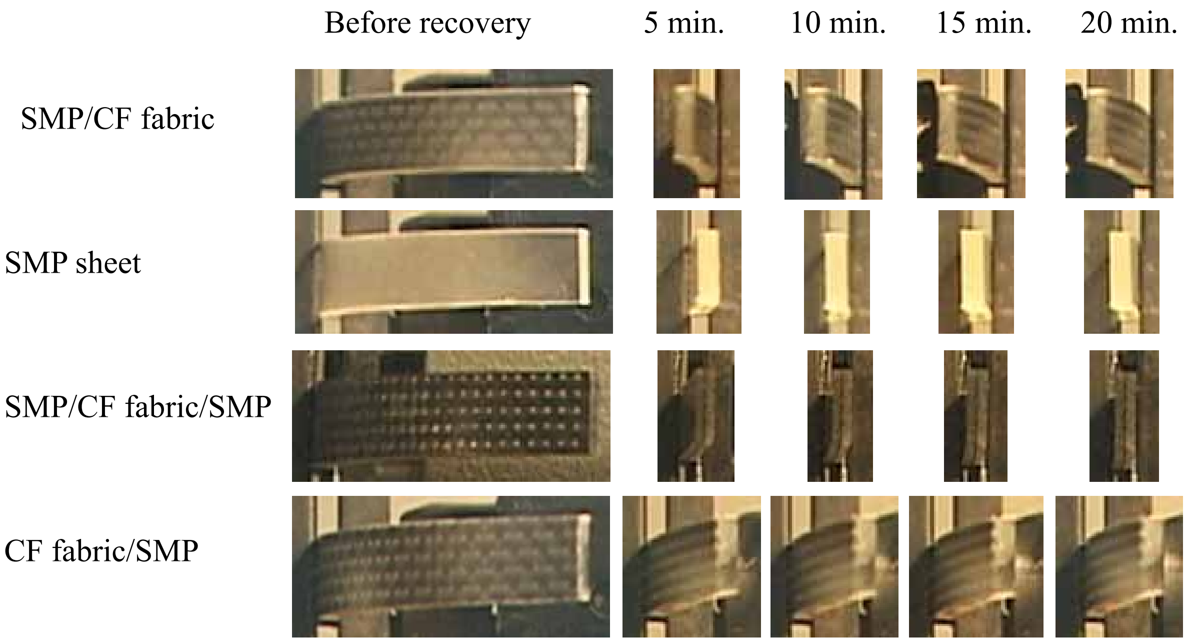


Fig.9. The bending recovery status without the self-weight influence in different recovery times at 65°C for SMP based laminates

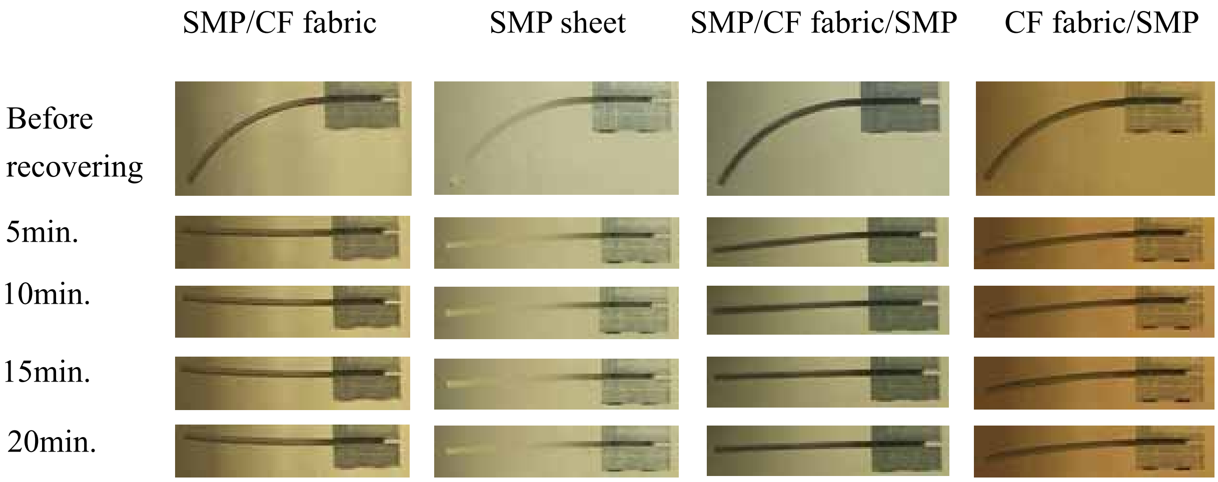


Fig.10. The bending recovery status with the self-weight influence in different recovery times at 65°C for SMP based laminates

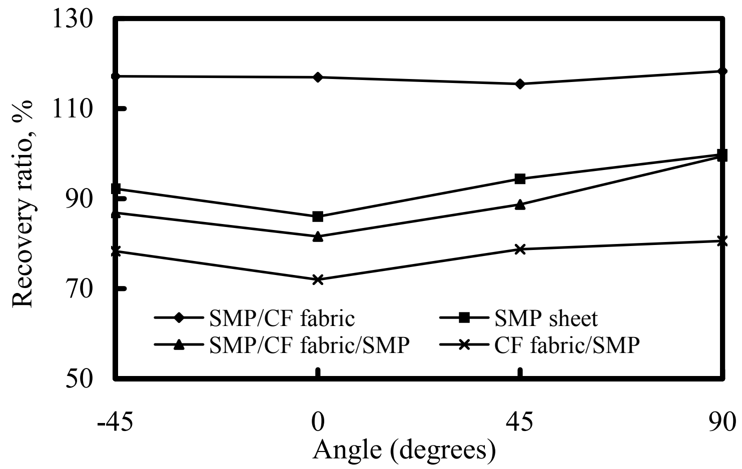
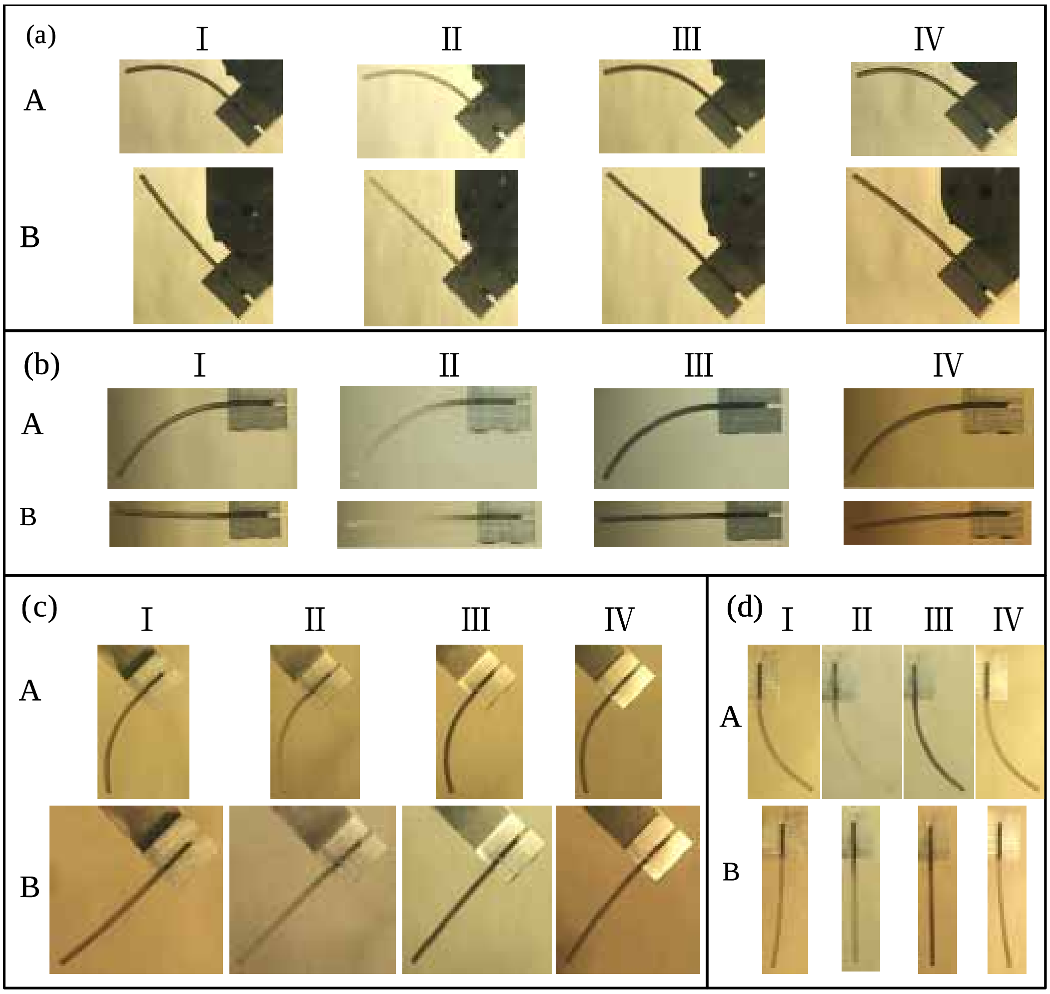


Fig.11. The relationship between the recovery ratio and the initial angle for SMP based laminates



(a) -45˚ (b) 0˚ (c) 45˚ (d) 90˚

Ⅰ: SMP/CF fabric Ⅱ: SMP sheet Ⅲ: SMP/CF fabric/SMP Ⅳ: CF fabric/SMP

A: Before recovering B: After recovering

Fig.12. The bending recovery status for different specimens under various initial angles

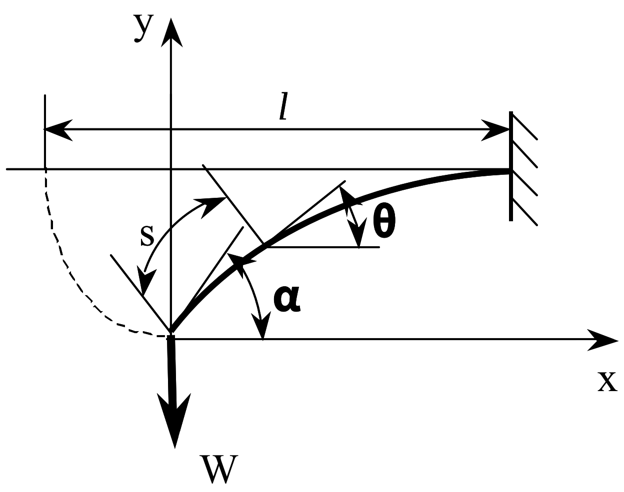


Fig.13. Bending stiffness $E_{CF}I_{CF}$ for CF fabric prepreg

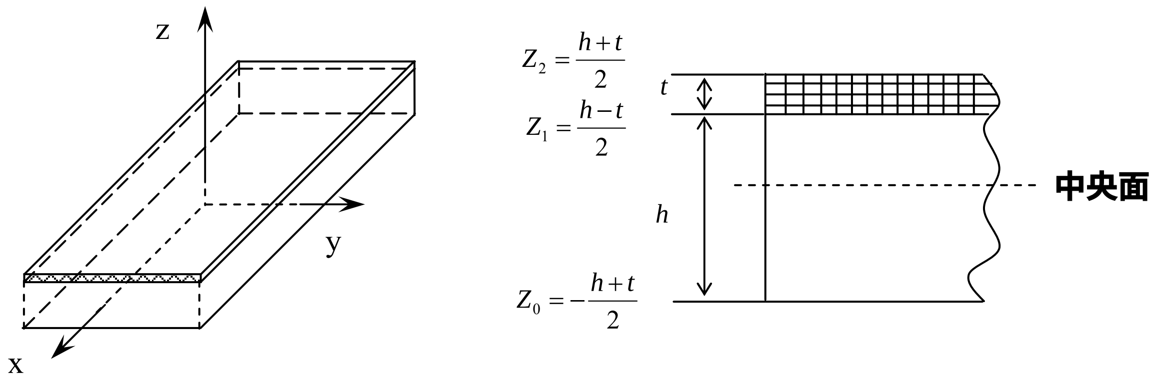
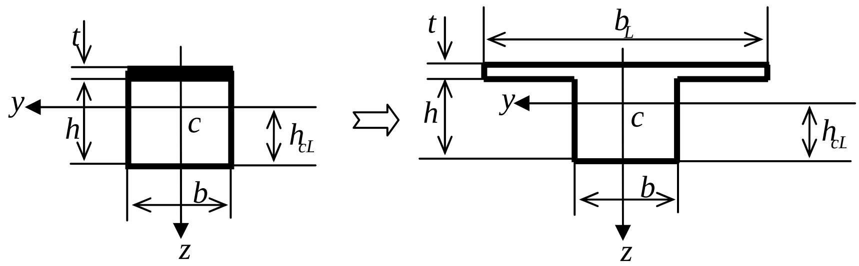


Fig.14. Cross section for SMP based laminates



(a) (b) equivalent section

Fig.15. Equivalent cross section of SMP based laminates

Table Ⅰ Geometrical and mechanical parameters

Thickness of the CF fabric prepreg	t	0.45 mm
Thickness of SMP	h	2.05 mm
Young's modulus of SMP at 15°C	E_{SL}	1630 MPa
Young's modulus of SMP at 65°C	E_{SH}	37.9 MPa
Poisson's ratio of SMP at 65°C	v_x	0.5
Curvature of middle surface at 65°C	k	0.0173

Table II Bending recovery ratio for SMP base laminates

Specimens	 SMP/CF fabric	 CF fabric/SMP	 SMP/CF fabric/SMP
Calculated result	129.72	68.71	99.99
Experimental result	119.12	77.72	99.53
Error	8.9%	-11.6%	0.5%