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A Comparative Experiment for the Analysis of Microwave and Thermal Process Induced Strains of Carbon Fiber/Bismaleimide Composite Materials

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Abstract: Carbon fiber reinforced bismaleimide composites provide many outstanding properties and are widely used in aerospace applications. However, cure-induced strains are present in virtually all composites that severely impact on the whole service lifecycle of composite components. This paper will demonstrate that the cure-induced strains can be drastically reduced in fiber/ bismaleimide composites using the microwave curing process. Nearly 95% reduction of cure-induced strains has been achieved compared with the conventional thermal heating process. The microwave manufacturing cycle for composites was only 36% of the thermal processing cycle. When using the microwave process, the spring-in angle of an L-shaped part was reduced by about 1.2^{o} compared by using thermal heating.

Key words: C. Residual stress, E. Microwave processing, A. Polymer-matrix composites (PMCs).

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1. Introduction

Carbon fiber reinforced bismaleimide (BMI) composites are widely used in aerospace industry, as the higher fracture toughness, better damage tolerance and longer fatigue life than carbon fiber composites with epoxy matrices. The traditional autoclave processing technology provides aerospace-grade composites with high strength and low defections, but the serious deformation causes big problems for further development [1].

Manufacturing induced stresses and deformation lead to intensive assembly effort or may even result in costly rejection of parts. The strong demands drive the aerospace industry to searching new curing technologies to replace the traditional thermal processing. Microwave curing technology, as one of the out-of-autoclave processes, is attracting more attention in solving those problems [2].

Microwave curing processes of advanced composite materials have been tried and studied by many researchers. Chou and Thostenson [3] and [4] first reviewed the possibility of microwave curing composite materials, and studied the fundamentals of microwave theory and applications of microwave heating process, especially the fiber-reinforced composites. Other researchers [5], [6] and [7] developed microwave processing of cross-ply carbon fiber/epoxy composites, experiments showing that under microwave field, thermosetting and thermoplastic composite materials can realize uniform curing and good quality. It was reported earlier by our research team [8] that, comparing microwave and thermal heating process, the temperature difference in laminates using modified microwave curing technology was reduced by 60%. Hunyar and Feher [9] developed autoclave-free systems with microwave heating techniques for composite processing, by using a special structure of microwave resonator. As mentioned above, the microwave curing procedure and difference with thermal processing have been addressed. Whereas, the possibility of microwave curing to reduce the deformation of composites still yet to be investigated.

In this paper, innovative microwave processing technology was tested in order to reduce the cure-induced strains and deformation of carbon fiber reinforced BMI composites. An investigation questionnaire was implemented in aerospace enterprises to statistically analysis the importance of difference influence factors. Fiber Bragg Grating (FBG) sensors were used to monitor the strains in real time. On account of the special forming mechanism of microwave energy, a quick curing process was investigated. Finally, the mechanisms of cure-induced strains during microwave curing were analyzed and the dimensional accuracy of an L-shaped carbon fiber reinforced BMI composite part was measured using a Coordinate Measuring Machine (CMM).

2. Industrial Investigation

For the purpose of reducing the deformation of advanced composite materials, the influencing factors of cure-induced strains have been studied by many previous researchers [10], [11] and [12]. For the reduction of composites' deformation, a comprehensive understanding of different mechanisms involved in practice is essential. A questionnaire-based investigation was carried out in Chengdu Aircraft Industry Group Co., and the results were shown in Fig. 1. A total of 19 questions have been answered by telephone interviews of engineers and technical managers.

Five influencing factors have been investigated and the horizontal axis means the percentage of different factors contribution to deformation. It can be seen in Fig.1 that the engineers in aerospace industry considered that the fiber/matrix Coefficient of Thermal Expansion (CTE) mismatch was the utmost factor which impacts the manufacturing stresses of thermoforming. The heat transfer and convection in composite materials were the inherent disadvantages of conventional thermal curing, hence the fiber/matrix CTE mismatch always exist during thermal forming. The authors proposed a new microwave curing method which can penetrate into composite and primarily heat the carbon fiber. Thus, the carbon fiber and resin CTE mismatch may be improved.

3. The Experiment Carried Out

3.1. Sample preparation and devices used

The unidirectional prepreg used in this experiment is combined with T700 carbon fiber and toughened bismaleimide resin. This kind of material is typically used for the production of high performance composite structures in aerospace industry. Other materials used in the experiment were purchased from supplier Airtech.

The experimental test showed that the 0^{o} plies composite laminates had a significant curvature than other laying forms [13]. Base on this results, the composite laminate sample's stacking-sequence was $[0_5]$s, and its dimensions were 100mm in length, 100mm in width and 1.5mm in thickness. To improve the reliability of the experimental results, 10 samples were tested for each process. Both of the thermal and microwave curing profile of carbon fiber/BMI prepreg were set at 130 °C for 30 minutes and then the temperature were held at 200 °C for 120 minutes. The heating rate was 1.5 °C/min and cooling rate was 1.3°C/min. Metal materials are known to reflect microwave energy, thus a high strength silicon tool was manufactured, with low coefficient of thermal expansion ($4.5\times10^{-6}/^{o}C$), excellent surface roughness (0.008μm) and thermal stability.

A high performance octahedron microwave oven was designed and manufactured by the authors' research team, as shown in Fig. 2 (b). The fiber optic fluorescence sensor was employed to measure the curing temperature and the electrical conductive rubber was connected with oven door to achieve electromagnetic shielding. This construction can guarantee a more even electromagnetic wave distribution than regular oven by multiple reflections and splitting. The schematic of microwave curing and strain measuring process was shown in Fig. 2 (a). A closed-loop temperature control system was applied to continuously adjust the microwave power. The strain measuring process was implemented by using FBG sensor and optical sensing interrogator.

3.2. Elimination of carbon fiber arcing

Previous work of the authors [8] and Bhudolia's team [14] identified that microwave curing composite may cause arcing of the carbon fiber bundles. This phenomenon is related to conductor gaseous breakdown in electromagnetic fields. The carbon fiber gaseous breakdown voltage can be described by Townsend's breakdown formula [15] and [16]:

$$V_b = \frac{Bpd}{\ln[Apd / \ln(1+1/\gamma)]} \tag{1}$$

Where p is environment pressure; d is distance between two points; and γ is the secondary ionization coefficient. The calculation is equation (2). Constant A is the saturation ionization, and B is related to the excitation and ionization energies.

$$\frac{1}{\gamma} = \exp(Ad) - 1 \tag{2}$$

Based on this theory, as the distance between the carbon fiber can hardly be controlled, the carbon fiber gaseous breakdown was eliminated by reducing the parameter p (pressure), constant A and B. By applying high vacuum pressure (0.5Pa) and sulfur hexafluoride gas (low A and B), this process finally allowed the microwave curing of carbon fiber/BMI composites without arcing and had a better overall performance.

3.3. In-situ measurement of cure-induced strains

The essential structural characteristics of FBG sensors determine the inescapability of double sensitive on temperature and stress when monitoring the cure-induced strains [17] and [18]. A glass capillary is used to pack the FBG for the measuring of the temperature variation. By this means, the Bragg grating will not contact the materials. The glass capillary has the same physical properties with the FBG sensors. Therefore, the process-induced strains of carbon fiber/BMI composite part can be accurately measured in the thermal and microwave environment.

3.4. Dimensional accuracy measured using a Coordinate Measuring Machine

In the aerospace industry, L-shaped composite parts all have serious deformation. Therefore, in this experiment, an L-shaped carbon fiber/BMI reinforced composite part with ply sequence $[0_5]_s$ was processed by thermal and microwave cuing technologies, in order to verify the correctness and applicability of the experimental results. A Mistral 070705 Coordinate Measuring Machine (CMM) was used to measure the dimensional accuracy of two composite parts.

4. Discussion of Results

4.1 Analysis of cure-induced strains

Fig.3 shows the in-situ measuring results of carbon fiber/BMI composite samples by microwave curing and conventional thermal processing. Fig.3 (a) and (b) are the curing temperature and strain of composites during thermal processing and microwave curing respectively. Both of those curves have the same variation tendency. The strain changes in the laminate are also similar to the previous research by Kim et al [19]. On account of the shorter curing time to reach the maximum curing percentage which was observed in the microwave cured composites than thermal curing by other researchers [14] and [20]. Faster microwave curing cycle was experimented to measure the cure-induced strains. The results were shown in Fig.3(c). The quick process was heating to 130 °C on 5°C/min, dwell 30 minutes and then held at 200 °C for 80 minutes, then naturally cooling down to room temperature. Differential Scanning Calorimeter (DSC) was used to measure the cure degree of composite after microwave processed. Both of the composite parts manufactured by quick microwave curing and thermal heating exhibited complete curing.

In order to analyze the strain with different distinctive characteristics, the curves were divided into seven Stages. In the first Stage, the FBG sensors suffered from the vacuum pressure from outside. As the temperature rose, the resin molecular motion increased while viscosity was decreasing, vacuum pressure undertaken by carbon fibers were transferred on to the FBG sensor. At the same time, the carbon fiber contracted with warning up as the negative CTE (-0.4 ×10-6/^{o}C). Hence, the compressive strain occurred during this phase. Stages two and four were the isothermal process of BMI composite, the curves varies within a small range. During the first dwell temperature, the strains generated from resin flow caused by fiber distortion and tool-part interaction, as the resin still not solidified, the strains could be changed. In Stage three, the matrix resin started to cure and the fiber/matrix CTE mismatch emerged. Then the shrinkage appeared at second dwell temperature, Stage four. Meanwhile, the strains in the composite material created by previous Stages were locked as the resin solidification. In the cooling Stages, the strains in the composite released and new strains generated. In the case that the composite CTE is large than silicon tool for one order of magnitude, dramatic compressive strains made the curves appeared a sharp decline. After a few moments, the curves increased to positive strains very quickly. This phenomenon can be explained by the relationship between frictional force and relative slip, the fraction between composite and tool changed from static friction to dynamic friction. At stage six, the strain was slowly eliminated along with the decreasing of temperature. Stage seven is the demoulding condition, vacuum bags were dismantled and composite samples were put on a platform with no constraint. The sharp peak in this stage was produced by distraction of external force.

The reason of sharp peaks monitored at the heating up Stage of quick microwave process may induced by the tension strain of silicon tool. Due to the temperature changed from heating up to dwell, the carbon fiber stopped shrink at the end of first and third Stages in Fig. 3(c). However, the temperature of silicon tool still rapidly rising and tension strain of silicon tool impose on the composite. Therefore the sharp peak was monitored at the end of those Stages. We considered this phenomenon has certain contribution to reduce the cure-induced strains. The quick heating up also lead to the air temperature around the composite much lower than the composite sample, temperature fluctuation was occurred during the second isotherm Stage.

It was clearly observed that three different processing technologies showed almost the same strain variation tendency. In detail, the microwave cured samples released more strains during cooling down. In order to analysis the relationship between different procedure and cure-induced strains, the statistical graph of different Stages with standard deviations are exhibited in Fig.3(d). Five important Stages were compared, including two dwell Stages - cooling down and demoulding. Different color lumps represent the different Stages and the strain variation trend is shown in the curve. The measuring errors and the difference of cure-induced strains in difference areas were included in the standard deviations by multiple experiments. As shown in Fig. 3(d), the strains generated by microwave curing were close to thermal process during Satges two and four. However, the strains in cooling down and demoulding Stages were much lower than the thermal processing. The composite part dimensional accuracy was determined by the residual strain after demoulding, so the final strain of the seventh Stage responsible for the composite property and quality. Moreover, the microwave curing reduced the residual strains to about -5 $\mu\varepsilon$, compared with -97 $\mu\varepsilon$ of thermal curing. Nearly 95% reduction of cure-induced strains has been achieved. Meanwhile, the quick microwave manufactured composites can be taken out from oven when 200 °C dwell ended and is cooled down outside (the air temperature in the oven is 30°C). The process time is cut to three hours, only about 36% of conventional thermal forming.

The strains mostly induced by fiber/matrix CTE mismatch, tool-part interaction and shrinkage were locked inside composite materials. The cooling down and demoulding Stages were responsible for the release of strains generated and locked in the previous Stages. Under the same conditions of curing temperature procedure and tool materials, the tool-part interaction was exactly the same. Therefore, the microwave curing technology reduced the fiber/matrix CTE mismatch during processing. The schematic diagram was shown in Fig.4.

Considering the previous work of microwave curing composite [5], the carbon fiber bundles have a higher microwave absorbing ability than resin. The dielectric loss tangent of carbon fiber and resin are 2.5 and -0.04. The negative dielectric loss tangent of resin may due to the dielectric loss is too small. Carbon fiber was firstly heated during microwave curing, then the thermal was transferred to resin matrix. Therefore, the carbon fiber can be slid into the resin before the solidification of the interface between fiber and resin. Accordingly, the carbon fiber/matrix CTE mismatch was reduced, as shown in Fig.4. Further in-depth research on this mechanism is to be conducted in the authors' on-going research work.

4.2 The dimensional accuracy of L-shaped composite parts

In order to verify the measuring results of strains during microwave curing, two L-shaped carbon fiber/BMI composites were manufactured by both quick microwave processing and conventional thermal curing methods. The normal vector of each data point was calculated, and then the scattered data points were compensated to eliminate the influence of probe diameter. Through analysis of the data dots in CAD software, the spring-in angle and corner curvature were measured, as shown in Fig.5.

As shown in Fig.5, the spring-in angles of two composite parts were calculated by measuring the equally spaced data points' lines. Sequentially from one side to another, the angle of two parts were marked as $\alpha_1, \alpha_2, \alpha_3$ and $\beta_1, \beta_2, \beta_3$. The results of measurement were exhibited in Table 1. Both L-shaped parts have a good uniform of angle distribution and the difference is obvious. The quick microwave cured composite part owns less spring-in angle than the thermal cured part. Also, the microwave cured part fitted the forming tool very well. About 1.2 ^{o} spring-in angle difference was measured between the microwave cured composite part and the traditional thermal cured part.

5. Conclusion

Microwave and traditional thermal cure induced strains of carbon fiber reinforced bismaleimide composites were analyzed in this paper. In-situ strains monitoring of composite samples were accomplished by applying fiber Bragg grating sensors. The carbon fiber arcing phenomenon in microwave field was discussed. Experimental results shown that the microwave cured composite parts have much lower cure-induced strains than traditional thermal processed parts, and the curing time is cut to 36% of thermal forming. The decreasing mechanisms of strains have been analyzed. For the purpose of verifying the monitored results of cure-induced strains, the dimensional accuracy of L-shaped parts have been measured using a coordinate measuring machine. The spring-in angle of a microwave energy manufactured L-shaped part was lower than a thermal heated part for about 1.2^{o}. The experimental results have big potential applications in high performance aerospace structures.

Acknowledgements

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Figure and Table Captions

Fig. 1. Influence factors of carbon fiber/BMI composites' deformation.

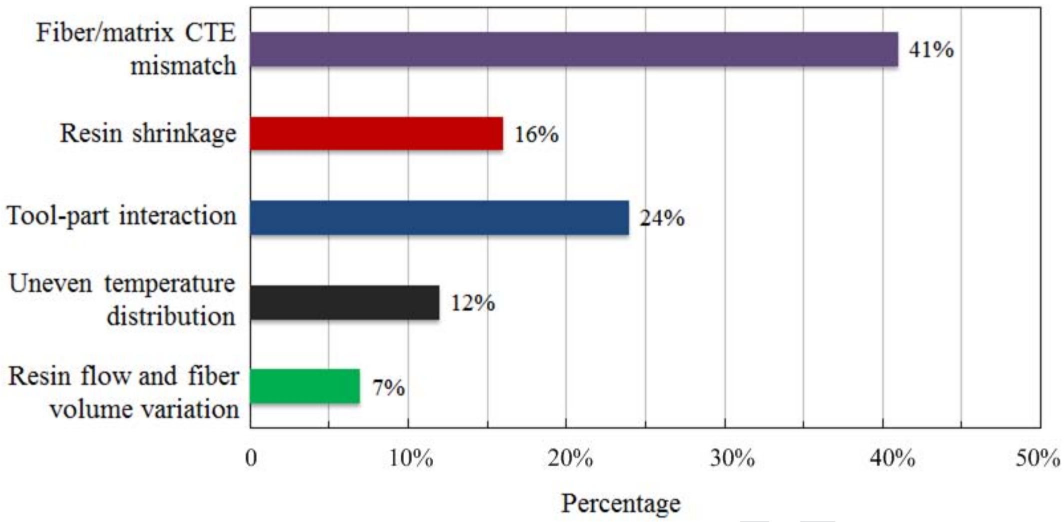
Fig. 2. The schematic of microwave curing and devices employed; (a) schematic of microwave curing and strain measuring; (b) the high performance octahedron microwave oven.

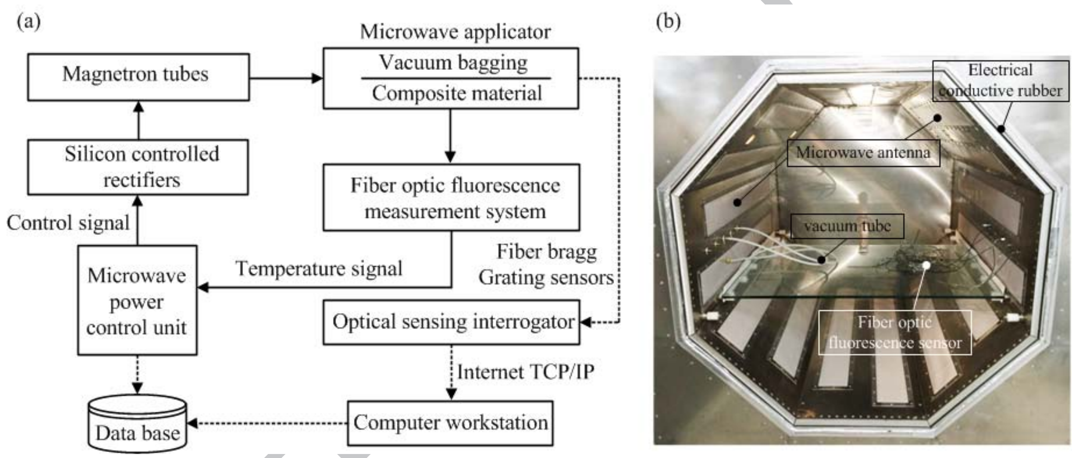
Fig. 3. In-situ measuring of composite curing induced strain and statistical graph with standard deviations: (a) microwave curing; (b) thermal curing; (c) quick microwave curing; and (d) statistical graph of different phases with standard deviations.

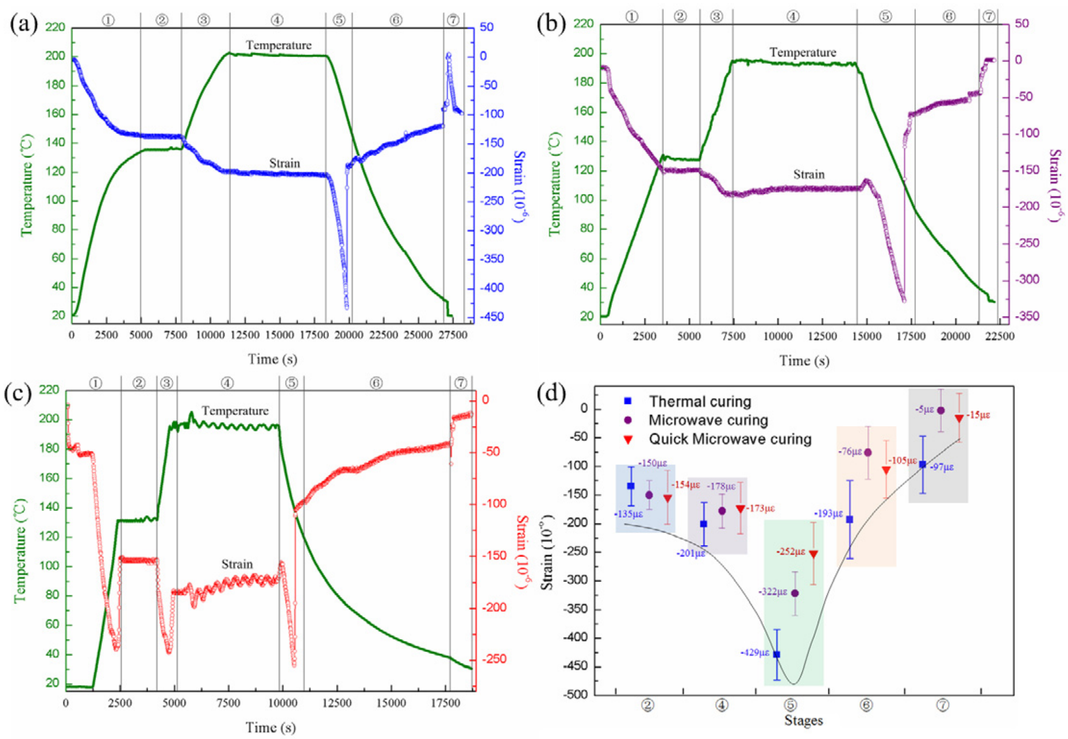
Fig. 4. Schematic diagram of cure-induced strain generation during thermal and microwave curing.

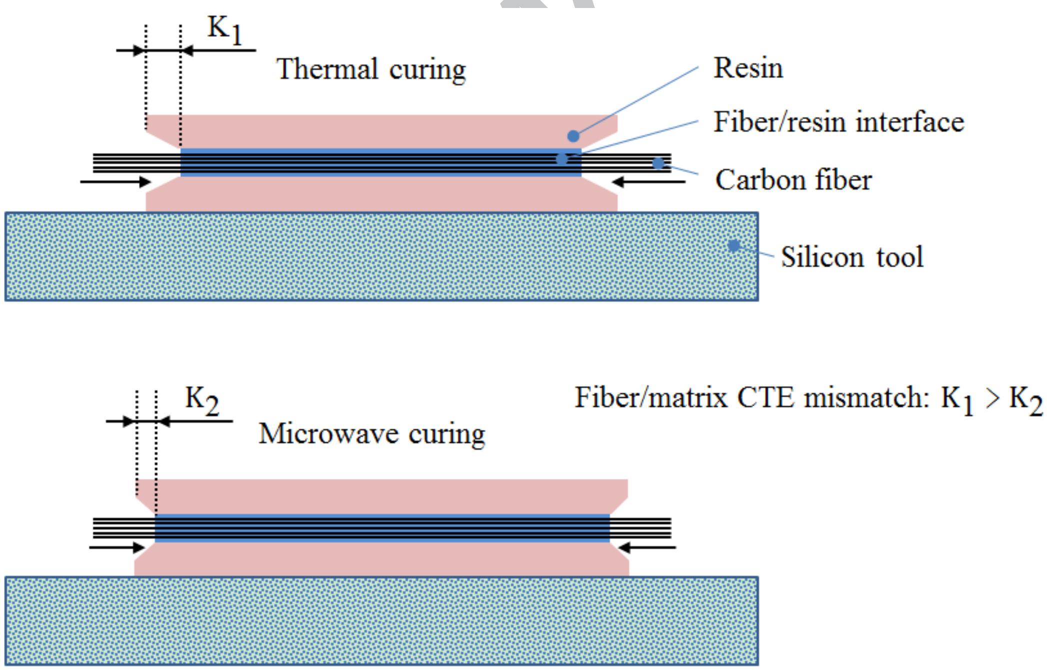
Fig. 5. Spring-in angle and corner curvature of L-shape parts: (a) data points of quick microwave cured part; and (b) data points of thermal heated part.

Table 1
Comparison of spring-in angle of composite parts manufactured by quick microwave and thermal processing.


Fiber/matrix CTE mismatch
Resin shrinkage
Tool-part interaction
Uneven temperature distribution
Resin flow and fiber volume variation
41%
16%
24%
12%
7%
0
10%
20%
30%
40%
50%
Percentage


(a)
Microwave applicator
Magnetron tubes
Vacuum bagging
Composite material
Silicon controlled rectifiers
Fiber optic fluorescence measurement system
Control signal
Microwave power control unit
Temperature signal
Fiber bragg Grating sensors
Optical sensing interrogator
Internet TCP/IP
Data base
Computer workstation
(b)
Electrical conductive rubber
Microwave antenna
vacuum tube
Fiber optic fluorescence sensor


(a)
(b)
(c)
(d)
Temperature
Strain
Temperature (℃)
Strain (10⁻⁶)
Time (s)
Thermal curing
Microwave curing
Quick Microwave curing
Stages
-135με
-150με
-154με
-201με
-178με
-173με
-429με
-322με
-252με
-193με
-76με
-105με
-97με
-5με
-15με


K1
Thermal curing
Resin
Fiber/resin interface
Carbon fiber
Silicon tool
K2
Microwave curing
Fiber/matrix CTE mismatch: K1 > K2

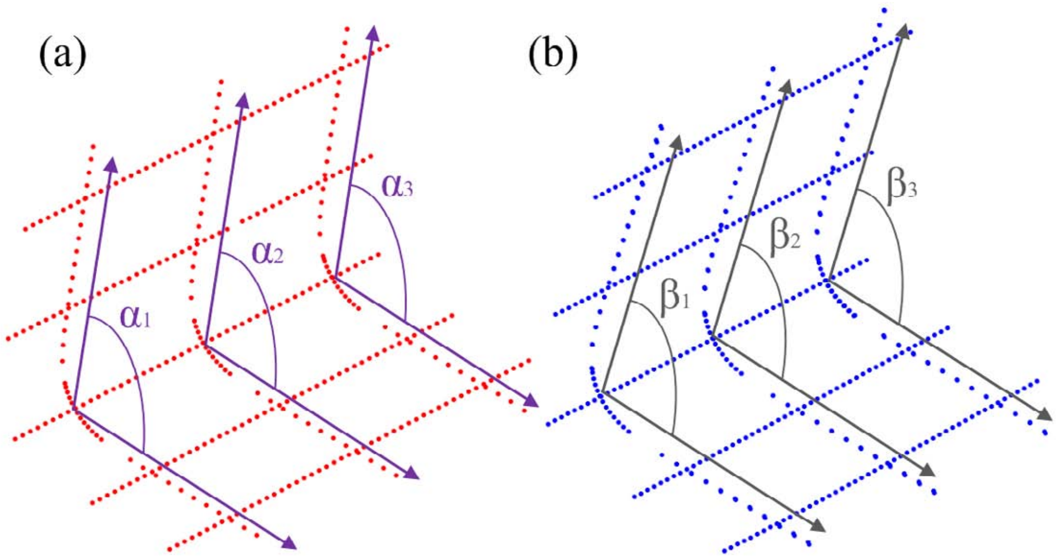

(a)
α1
α2
α3
(b)
β1
β2
β3

Table 1
Comparison of spring-in angle of composite parts manufactured by quick microwave and thermal processing.

	α_1 and β_1	α_2 and β_2	α_3 and β_3
Quick microwave cured part	90.24^o	90.10^o	90.63^o
Thermal cured part	88.33^o	88.97^o	89.96^o