**Summary:** The effects of temperature cycle and process gap on the thermal expansion pressure and resin pressure were investigated. It indicates that the process gap influences magnitude of expansion pressure and the moment of generating pressure. The temperature cycle with lower temperature plateau can effectively reduce the gradient inside the mold.

1 General Introduction

As is well known, processing of high-performance composite structures in the aviation industry is usually done using autoclaves, since the devices can apply an appropriate amount of heat and pressure under a vacuum for cure and consolidation of composite parts. However, it is of great difficulty to provide appropriate compaction pressure to the composite parts with a three-dimensional complex structure such as tubes, inserts, ribs, etc. Thus, voids, delaminations and fiber bridging, which significantly reduce the mechanical properties of composites, are often observed in these structures. As to overcome these problems, thermal expansion molding method was introduced to apply uniform compaction pressure over a complex surface [1].

As the thermal expansion pressure is crucial for processing composites during process, there are a lot of researches focused on the control of thermal expansion pressure of the silicone rubber mold[2,3,4,5]. However, the actual expansion pressure exerted on prepreg stack can not be directly tested. Thus, there is a lack of deep understanding on the relationship between thermal expansion pressure and process parameters, i.e. process gap and temperature cycle. Consequently, it is difficult to analyze the resin pressure inside prepreg stack, which has a significant effect on controlling void defects of final composites[6,7].

In order to obtain the fundamental understanding on the relationship among process parameters, thermal expansion pressure and resin pressure during thermal expansion molding process, carbon fiber/bismaleimide resin composite laminates were produced using a designed processing mold. By means of the mold and a resin pressure online measuring system [8], the thermal expansion pressure generated by silicone rubber mold and resin pressure inside prepreg stack were measured. In addition, the effects of process gap and temperature cycle on thermal expansion pressure and resin pressure were investigated. Finally, qualities of the laminates cured using different process gaps were studied.

2. EXPERIMENT

2.1. Thermal Expansion Pressure Investigation

2.1.1. Measurement of thermal expansion pressure

In this study, a processing mold was designed to measure the thermal expansion pressure and the schematic diagram is shown in Fig. 1.

The mold is heated by four heating rods which are located in upper and lower molds respectively, and the temperature is adjusted by a temperature controlling system. The process gap can be adjusted by the thickness of gaskets.

Aircast 3600, supplied by Airtech International, Inc., was selected to cast silicone rubber mold, and its dimension is 15cm×15cm×4cm.

Fig. 1. Cross section of the processing mold.

During the measurement of thermal expansion pressure, the processing mold was clamped by the upper and lower cross heads of a CMT550 universal testing machine. The cross heads were set in “no displacement” mode and load resulted by the thermal expansion should be recorded.

The carbon fiber/bismaleimide resin unidirectional prepreg tapes with the resin mass fraction of 37% were cut into pieces with dimension of 15×15 cm². The 16-layer prepreg stacks were laid up with cross ply and then bagged with nonporous Teflon film to prevent resin bleeding. Finally, the laminates cured in the processing mold.

Two temperature cycles were selected for the cure of laminates. Cycle one: the temperature was directly increased from room temperature to 160°C at 2°C/min and then held for 2 hours, and last decreased to room temperature. Cycle two: the temperature was first raised from room temperature to 130°C at 2°C/min and then held for about 1 hour, and then increased to 160°C and kept constant for 1 hour. At last, the temperature decreased to room temperature. All the laminates were postcured in an oven at 190°C for 2 hours for further investigation.

2.1.2. Blank test

It is necessary to conduct a blank test because of thermal expansion of metal mold. During the process, only the load reduced by thermal expansion of processing mold was measured.

As a result, the thermal expansion pressure of silicone rubber mold (P) at testing temperature (T) can be obtained by the following equation:

\[ P = \frac{F - F_{\text{blank}}}{S} \]  

Where \( F \) is the load obtained during the measurement of thermal expansion pressure; \( F_{\text{blank}} \) is the measured load during blank test; \( S \) is the contact area between silicone rubber mold and prepreg stack.

2.2. Measurement of Resin Pressure

The resin pressure measurement was conducted by a pressure sensor. The schematic of the measuring system is shown in Fig. 2.


Fig. 2. Schematic of hydrostatic resin pressure measuring system.

The accuracy and dynamic response of the measuring system can well meet the requirements of resin pressure measurement during cure cycle[8].

During the measurement, the sensor probes were embedded at center and edge regions, as the circles schematically shown in Fig. 3.

Fig. 3. Schematic of positions for resin pressure testing probes placed in the prepreg stack.

2.3. Measurement of Temperature

The temperature distribution inside silicone rubber mold and prepreg stack during cure cycle was tested by a temperature field measuring system. The thermocouples were embedded into different positions of the silicone rubber mold (see the circles in Fig. 4).
cross sections were wet mold and expansion pressure generated on pressure of resin, the restricted volume of gel should. The cured laminates were cut into small pieces, and the cross sections were wet ground with successively finer silicon carbide paper, and wet polished with chromium oxide. The polished sections were then observed using the optical microscope (Olympus BX51M).

3. DESIGN OF PROCESS GAP

As the temperature increases, the volume of silicone rubber mold expands. At the gel temperature of resin \( T_{gel} = 160 \, ^\circ C \), the volume of the silicone rubber mold without restriction \( (V) \) should be calculated using following equation:

\[
V = V_0 \left[ 1 + \alpha \left( T_{gel} - T_0 \right) \right]
\]  
(2)

Where \( V_0 \) is the original volume of silicone rubber mold at room temperature \( (T_0) \), \( 970.6 \times 10^{3} \text{mm}^3 \); \( \alpha \) is the volume expansion coefficient of silicone rubber, \( 5.6 \times 10^{3} \text{C}^{-1} \).

As the thermal expansion pressure generates under restriction condition, the restricted volume of the silicone rubber has a significant effect on the expansion pressure and the relationship is addressed:

\[
P_{gel} = K \frac{\Delta V}{V}
\]  
(3)

Where \( P_{gel} \) is the thermal expansion pressure of silicone rubber mold at gel temperature \( (T_{gel}) \); \( K \) is the bulk modulus of silicone rubber, \( 722 \text{MPa} \); \( \Delta V \) is the restricted volume of silicone rubber mold.

As the processing mold cavity and rubber mold are cube, the process gap \( (L_{gap}) \) can be obtained according to equation 4.

\[
L_{gap} = \frac{V - V_0 - \Delta V}{S}
\]  
(4)

As a result, the appropriate process gap is 3.5mm. In addition, the process gap of 2.9mm and 4.0mm were chosen for comparison.

4. RESULTS AND DISCUSSION

4.1. The Effect of Temperature Distribution on the Expansion Pressure

Because of the low thermal conductivity \( (0.21 \text{W.m}^{-1}.\text{K}1) \) of silicone rubber mold, temperature gradient inside silicone rubber mold inevitably exists during ramp stage of the temperature cycle. The temperature at different positions and expansion pressure were tested and the results are shown in Fig.5 and Fig.6, respectively.

Fig.5. Measured temperature field and expansion pressure during temperature Cycle One. Fig.5 shows the temperature distribution inside the rubber mold and expansion pressure generated by the rubber mold during temperature Cycle One. Once heating process begins, the temperature at the bottom of the rubber mold increases immediately. About 800 seconds later, the temperature at center of the mold starts to rise. As a result, it presents a significant temperature gradient in the thickness direction of the mold and the max difference between center and bottom is \( 35 \, ^\circ C \). However, almost no difference of the temperature between center and edge positions (see in Fig. 4) is observed. As the bottom region of the rubber mold reaches to \( 160 \, ^\circ C \), the temperatures at edge and center just reach to \( 140 \, ^\circ C \).

The volume expansion of the silicone rubber mold induced by the increase of temperature firstly fills the process gap. During this stage, no expansion
pressure generates. As the bottom of the mold reaches to 150°C, the expansion pressure starts to exert on the surface of prepreg stack. Then, it increases to 0.60MPa. As the heating rate decreases subsequently, the expansion pressure increases with a lower rate and finally reaches to 0.70MPa when the temperature of center region is 165°C.

In order to reduce the temperature gradient inside the silicone rubber mold, the temperature cycle with a lower temperature plateau (Cycle Two) was selected for investigation and the results are shown in Fig.6.

![Fig. 6. Measured temperature field and expansion pressure during temperature Cycle Two.](image)

Before the first plateau, great temperature variation between bottom and center forms and the max difference is 30°C. After this plateau, the temperature gradient inside rubber mold also exists. However, the difference obviously decreases and the max difference is about 10°C.

As the bottom of the mold reaches to 130°C, the expansion pressure starts to apply on the prepreg stack. Then, it increases to 0.60MPa when it reaches to 160°C. The lower temperature plateau reduces the temperature gradient inside the mold. In addition, because the prepreg stack is placed next to the bottom of the mold, it starts to be compacted at lower temperature.

### 4.2. The Effect of Process Gap on the Expansion Pressure

In order to investigate the effect of process gap on expansion pressure and resin pressure inside prepreg stack, the process gaps with different sizes (2.9mm, 3.5mm and 4.0mm) were chosen for the study and the results are shown in Fig.7.

![Fig. 7. Measured expansion pressure and resin pressures inside prepreg stack with different process gaps.](image)

As the temperature increases, the silicone rubber molds expand and the process gap generally decreases. During this stage, the silicone rubber mold can not be restricted by the metal mold. Accordingly, no expansion pressures generated and no resin pressures were measured. Once the temperature reached to approximately 80°C, the expansion pressure generated under the condition of 2.9mm process gap, and then immediately exceeded 1.0MPa. At the same time, the resin pressures inside the prepreg stack increased significantly. In addition, obvious resin pressure gradient occurred from center to edge and the resin was squeezed out.

In the case of 3.5mm process gap, expansion pressure starts to compact prepreg stack at about 120°C and rapidly increases to 0.65MPa as the temperature reaches to 145°C. Then, the temperature continues rising with a lower rate until it reaches to 160°C. Similar trend is observed for the curves of resin pressures. At the end of ramp of temperature cycle, the resin pressure increases to about 0.68MPa. During the whole process, the two curves of resin pressures at center and edge are almost overlapped. It indicates that the resin pressures uniformly distribute inside the prepreg stack. It additionally proves that silicone rubber mold can provide even compaction pressure on surface of the stack.

Focused on the process gap of 4.0mm, it is found that only 0.15MPa expansion pressure was induced by the silicone rubber mold when the temperature reached to 160°C, which was far less than the compaction pressure recommended by the prepreg supplier.
4.3. Defects of the Laminates

The micrographs inside the laminates processed using different process gaps were further observed and the results are shown in Fig.8.

(A) 2.9mm process gap

(B) 3.5mm process gap

(C) 4.0mm process gap

Fig.8. Micrographs of the laminates processed using different process gaps.

As extreme high compaction pressure was applied on the surface of the laminate processed using 2.9mm process gap, resin over-bleed from the prepreg stack. Resin starvation and poor bonding between layers occurred. As a result, delaminations formed during machining as shown in Fig.8(A). In the case of the laminate processed using 3.5mm process gap as shown in Fig.8(B), no defect was found inside. It is mainly attributed to the appropriate expansion pressure applied on the prepreg stack. On one hand, no obvious resin bleeding occurs and it results in proper consolidation of the laminate. On the other hand, it efficiently controls the formation of voids. Fig.8(C) indicates that poor consolidation and voids are discovered in the laminate processed using 4.0mm process gap. In this case, the low expansion pressure can not compact the prepreg stack effectively. In addition, the voids can be eliminated under the low level of resin pressure.

5. CONCLUSIONS

This paper establishes an efficient method to investigate the thermal expansion pressure and the resin pressure during thermal expansion molding process. The experimental results demonstrate that the process gap significantly influences magnitude of expansion pressure and the moment of generating pressure. In addition, the temperature cycle with lower temperature plateau can effectively reduce the temperature gradient inside the silicone rubber mold and can compact the prepreg stack at lower temperature. It provides guidance for optimization of process parameters during thermal expansion molding.

6. ACKNOWLEDGEMENT

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