

Co-curing process combining resin film infusion with prepreg and co-cured interlaminar properties of carbon fiber composites

Weiwei Xu¹, Yizhuo Gu¹, Min Li¹, Xuqiang Ma¹,
Dongmei Zhang² and Zuoguang Zhang¹

Abstract

In this article, the carbon fiber/epoxy resin matrix composite laminates were fabricated using a co-curing process combining resin film infusion (RFI) process with prepreg-autoclave process, called co-resin film infusion. A kind of unidirectional prepreg and its corresponding resin film were adopted. The compaction and defects of laminates cured by different processing were studied. Mode I and mode II interlaminar fracture toughness were adopted to evaluate the co-cured interlaminar properties of the co-curing laminates and were compared with those of laminates processed by the prepreg-autoclave process and the resin film infusion process. Moreover, the effects of lay-up type of prepreg part and resin film infusion part, isothermal dwell and epoxy tackifier of fiber preform were also studied. The results show that these factors have important effects on the processing qualities of the laminates cured by co-curing process, including resin-rich regions and voids, resulting in different interlaminar fracture toughness at the co-cured interface. Affected by the prepreg part and the resin film infusion part in the co-resin film infusion laminates, the mode I and mode II initial interlaminar fracture toughness of co-cured laminate lie between those of the prepreg laminate and resin film infusion laminate, and G_{IC} at crack propagation stage for co-resin film infusion laminate are higher due to fiber bridging and deflection of crack. These results have close relationships with the compacting structure of fibers in prepreg and resin film infusion parts and the interfacial bonding between the two kinds of fiber and the matrix.

Keywords

Carbon fiber composites, co-curing process, resin film infusion, prepreg, interlaminar fracture toughness

Introduction

Resin film infusion (RFI) is a low-cost composite manufacturing process for advanced composite structure with high mechanical performance. In RFI process, resin film is placed on mold surface with dry fiber preform, melts by heating and penetrates the fiber pre-forming body under vacuum pressure or external pressure, and then is usually cured in an autoclave. Gas inside the fiber preform is eliminated via the flow of resin film.^{1–3} Compared to prepreg technology, RFI has been considered a composite manufacturing process for large parts with high-cost reduction potential. The applications of RFI are mainly in aerospace field, such as AST-Composite Wing program carried out by NASA/Boeing and the pressure bulkhead of Airbus A380.⁴

Co-cured liquid composite molding (co-LCM), initially established by an American company named Northrop Grumman and developed in the project of composite affordability initiative by United States department of defense, is an integral forming technique

¹Key Laboratory of Aerospace Materials and Performance (Ministry of Education), School of Materials Science and Engineering, Beihang University, Beijing, P. R. China

²National Engineering and Research Center for Commercial Aircraft Manufacturing, Commercial Aircraft Corporation of China, Ltd, Shanghai, P. R. China

Corresponding author:

Weiwei Xu, XueYuan Road No.37, HaiDian District, Beijing, 100191, China.
Email: demon0926@126.com

for combining liquid molding process with prepreg curing process.^{5,6} It combines prepreg stack with dry fiber perform, subsequently the fiber perform is infused by liquid resin and finally integral composite product including prepreg part and LCM part is cured. The co-LCM technology improves the processability of complex structure and achieves the purposes of time-saving and cost-saving by means of reducing fasteners, assembling procedures and simplifying mold. For example, Kaps^{7,8} studied the combination of resin injection process and prepreg process, and suggested that the co-curing process had the potential to considerably reduce the production time of large assemblies for primary structural applications. In addition, the application of co-LCM in resin transfer molding (RTM), which is called co-RTM, has been developed to fabricate aircraft structures, such as vertical stabilizer. Considering that both RFI and prepreg processing are adopted using autoclave for aircraft composite structures, we propose a new idea of co-RFI, which co-cures RFI part with prepreg stack simultaneously for taking advantages of cost-effectiveness of RFI and high properties of unidirectional prepreg.

Obviously, two kinds of resins, the resin in prepreg stack and the resin for infusion, are utilized during co-LCM process, and it is one of main features for this co-curing manufacturing method. The compatibility of the two resins and its effects on the processing and mechanical properties of the co-cured composite need to be understood. Especially, the interface region between the prepreg part and the LCM part is formed via the interaction of the two resins, and the interface properties should be different from those in both individual parts. In addition, the compacting structures of fiber reinforcements in prepreg part and LCM part are usually different, and their effects on resin flow and composite consolidation at the co-curing interface are worth studying. However, the relevant investigations are seldom reported.

The work presented herein puts forward a developing co-curing process, named co-RFI, which combines conventional RFI process with prepreg-autoclave process. A carbon fiber/epoxy resin prepreg and its corresponding resin film were adopted to fabricate composite laminates using co-RFI process. The processing quality of the co-cured laminate was compared with those of RFI laminate and prepreg laminate. Furthermore, mode I and mode II interlaminar fracture toughness were measured to evaluate the co-cured interface between the prepreg part and the RFI part. The effects of lay-up type, temperature cycle and epoxy tackifier for fiber preform on the properties of co-RFI composite were studied. The results indicate some important issues for the application and development of co-RFI technology.

Experiment

Materials

A kind of epoxy resin film, named MTM 44-1, supplied by Advanced Composites Group Ltd (ACG) was used for this experiment. The resin film had 200 g/m² areal weight. T700SC (12k tow) unidirectional carbon fiber fabric with 200 g/m² areal weight (CFW-200, supplied by Jiangsu Tianniao High Technology Co. Ltd) was used as reinforcement in RFI process. HTS 5631 (12k tow)/MTM 44-1 unidirectional prepreg with 134 g/m² areal weight provided from ACG was used in the present study. Its resin weight fraction was 35% and the nominal cured thickness per ply of the prepreg was 0.125 mm. An epoxy tackifier produced by Beijing Institute of Aeronautical Materials was adopted for binding CFW-200 fabrics in RFI process. The tackifier is powder and can shape fiber preform under heating after being dispersed on the surface of fiber fabric.

Composite manufacturing

Prepreg-autoclave process. MTM 44-1 unidirectional prepreg laminate of [0°]₃₂ with 300 mm × 200 mm was cured with a Teflon film placed between the 16th and 17th layers to form a pre-crack for the measurement of interlaminar fracture toughness. The manufacturer's recommended cure cycle was applied in autoclave. A consolidation pressure of 0.4 MPa and -0.1 MPa vacuum were applied throughout the cure process and the temperature was increased from room temperature to 180°C at 2°C/min and maintained at 180°C for 2 h.

RFI process. The schematic diagram of the bagging procedure of RFI process is shown in Figure 1. The stack contained 20 layers of CFW-200 carbon fiber fabric plies in 0° direction and nine layers of resin film, which were laid-up on a single-sided mold tooling and then sealed in a vacuum bag. The assembly was transferred into an autoclave with the consolidation pressure of 0.4 MPa and -0.1 MPa vacuum.

The release film was used perforated to ensure entrapped air to be released. In addition, a Teflon film was placed in the mid-plane of the fiber stack as the initial crack for the measurement of interlaminar fracture toughness, as sketched in Figure 2.

In order to study the effects of processing conditions on the properties of composite fabricated using RFI, three types of processes were chosen, as described below.

1. Heat from room temperature (25°C in this study) to 180°C at 2°C/min and hold an isothermal dwell at 180°C for 120 min, which is the manufacturer's

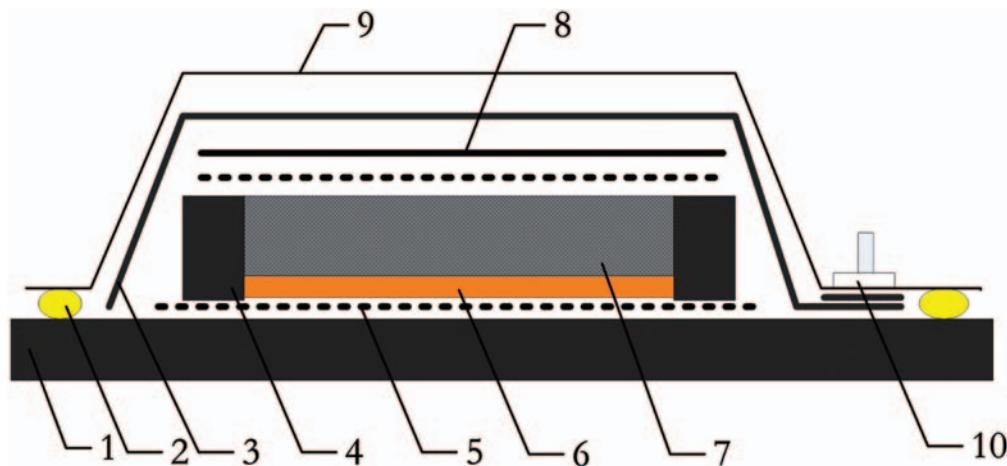


Figure 1. Schematic of the bagging procedure in RFI process.

1 – steel plate, 2 – sealant, 3 – breather, 4 – dam, 5 – peel ply, 6 – resin film, 7 – fiber fabric, 8 – release film, 9 – vacuum bag, 10 – vacuum valve.

RFI: resin film infusion.

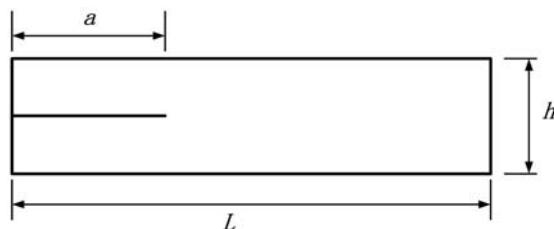


Figure 2. Sample geometry of interlaminar fracture toughness sample.

L = length, h = thickness, a = initial crack length.

- recommended cure cycle and is called Process A in this article.
- Heat from room temperature to 130°C at 2°C/min and hold an isothermal dwell at 130°C for 30 min and then increase to 180°C at 2°C/min with an isothermal dwell of 120 min. This condition is called Process B in this article.
 - Disperse 8 wt% tackifier on the surface of CFW-200 fabrics and maintain 5 min at 80°C to make the tackifier adhere to the fiber. The treated fabric was used to make fiber preform. Then, the same cure cycle as Process B was used to carry out RFI process. This condition is called Process C in this article.

Co-RFI process. Co-curing composite laminate, including 16 MTM 44-1 prepreg layers, 10 CFW-200 fabric plies and five layers of resin film, was laid up in 0° direction, and two kinds of lay-up types were designed (see Figure 3) to investigate the effect of lay-up type on the properties of composite laminates. One lay-up

type was putting the prepreg layers at the top of stack (above the dry fiber fabric) and is called prepreg/RFI, the other lay-up type was putting the prepreg layers on the bottom of stack (below the dry fiber fabric and resin film) and is called RFI/prepreg. In addition, a Teflon film was placed between the prepreg layers and the fabric plies as the initial crack for the measurement of interlaminar fracture toughness.

The co-RFI laminates were manufactured in an autoclave using Processes A, B and C, respectively. The bagging procedure was as same as the bagging procedure shown in Figure 1.

Testing

Resin viscosity. The viscosities of the resin film and tackifier were measured using a Gemini rheometer made by Bohlin Instruments. The rheometer was employed in parallel-plate configuration with a disc radius of 25 mm and gap size 0.6 mm. Data were generated with the disc oscillating at 1.0 Hz and 10 Pa stress. The dynamic temperature ramp was 2°C/min for the resin film and tackifier, and the isothermal temperatures were 120°C, 130°C, 140°C and 150°C for the resin film.

Mode I interlaminar fracture toughness. In this work, mode I interlaminar fracture toughness of different composite laminates was studied using the double cantilever beam (DCB) specimen according to American Society for Testing and Materials (ASTM) standard D 5528, wherein the specimen from the co-curing laminate was tested to obtain the interlaminar fracture toughness at the interface region between prepreg part and RFI part.

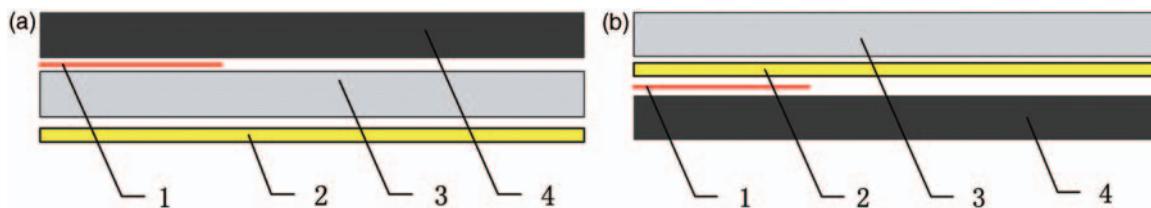


Figure 3. Schematic of two kinds of lay-up types: (a) prepreg/RFI stack and (b) RFI/prepreg stack.
1 – Teflon film, 2 – five layers of resin film, 3 – 10 carbon fiber fabric plies, 4 – 16 prepreg layers.
RFI: resin film infusion.

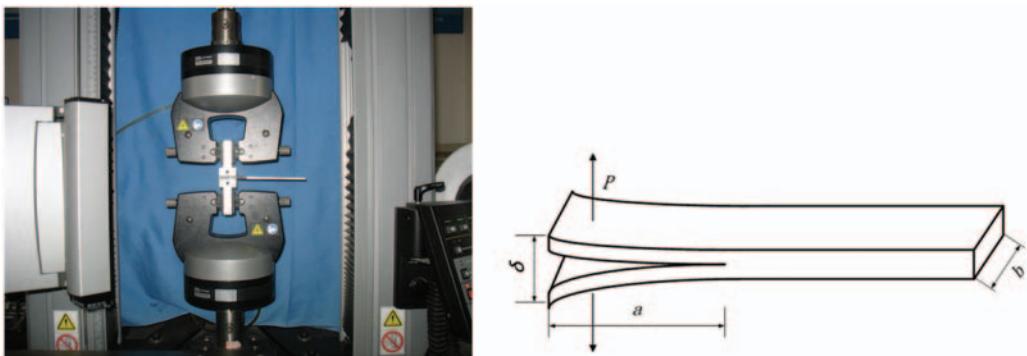


Figure 4. Schematic of DCB testing: a = crack length, p = Load, δ = displacement.
DCB: double cantilever beam.

For the DCB specimens of 25 mm in width and 150 mm in length, a non-adhesive insert (Teflon film) was inserted at the mid-plane of the laminate during lay-up to form an initiation site for the delamination. The testing was conducted on an Instron 5565 test apparatus at a displacement rate of 1 mm/min using a loading block, as shown in Figure 4.

Mode I interlaminar fracture toughness was calculated as follows:^{9,10}

$$G_{IC} = \frac{3P\delta}{2b(a + |\Delta|)} \quad (1)$$

where P is the load (N), δ is the load point displacement (mm), b is the specimen width (mm), a is the delamination length (mm) and Δ is the horizontal axis intercept from $a - C^{1/3}$ curve (shown in Figure 5). The compliance, C , is the ratio of the load point displacement to the applied load, δ/P .

The critical G_{IC} value was calculated as the average of the G_{IC} values at the beginning of delamination, $a = 50$ mm.

Mode II interlaminar fracture toughness. In this study, mode II interlaminar fracture toughness was investigated by using the end notched flexure (ENF) specimen according to Standard HB 7403-96. Five specimens were

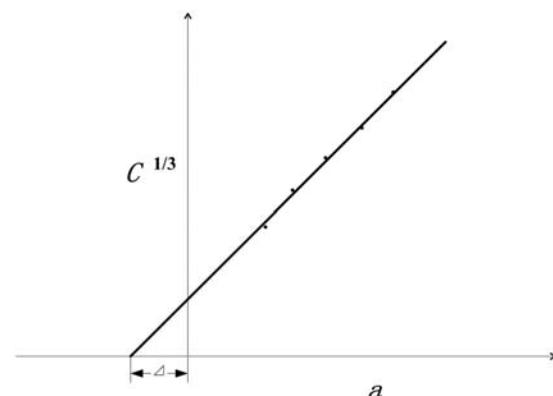


Figure 5. Schematic diagram of Δ .

tested from each panel, wherein the specimen from the co-curing laminate was tested to obtain the interlaminar fracture toughness at the interface region between prepreg part and RFI part.

The ENF specimens of 25 mm in width and 140 mm in length were employed to study the mode II fracture behavior. The specimens were intermittently loaded twice with sliding the loading and supporting points, where the initial crack length was set at 25 mm at the start of each loading process. The first loading was stopped after an increment of delamination crack

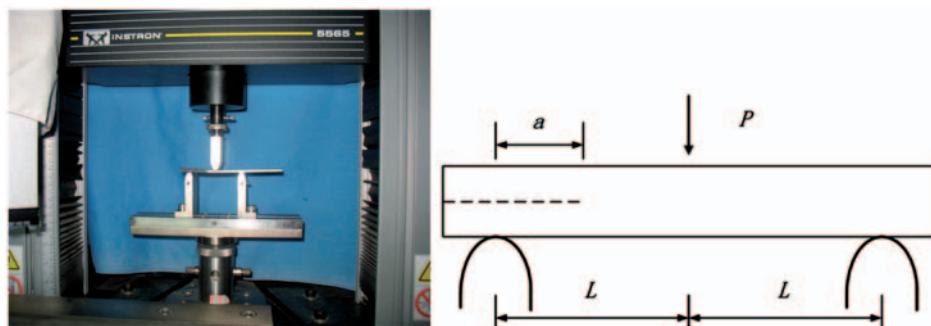


Figure 6. Schematic of ENF testing: a = the effective crack length, p = load, L = a half of the span length. ENF: end notched flexure.

growth of about 5 mm, with the span length 70 mm. The specimen was then reloaded with the span length 100 mm and unloaded when there was a deviation from linearity in the P versus δ curve. P recorded at the first loading was the crack initiation loading and the second one was the crack propagation loading.¹¹ The testing was conducted on an Instron 5565 test apparatus at a displacement rate of 1 mm/min using a three-point bend jig, as shown in Figure 6.

The mode II fracture toughness was investigated as the critical value of the energy release rate, G_{IIC} , on the basis of the linear fracture mechanics using the following equation:

$$G_{IIC} = \frac{9P\delta a^2}{2b(2L^3 + 3a^3)} \times 10^3 \quad (2)$$

where P is the load (N), δ is the load point displacement (mm), b is the specimen width (mm), a is the effective crack length (mm) and L is the half-span length (mm).

The initial G_{IC} value was calculated as the average of the G_{IC} values using the crack initiation loading of all the available specimens for each specimen group.

Processing quality observation

The processing qualities of cured laminates using different processes were characterized by optical micrograph and laminate thickness.

In this study, microscopic observation was used to investigate the defects inside laminates. The specimens from cured laminates were mounted in an epoxy resin, ground and then polished. The polished cross-sections were observed using Olympus BX51M optical microscope. The void contents of the laminates and the co-cured interfaces were also studied through the digital microscopy and image analysis. For measuring the void contents of the co-cured interfaces, we only chose certain regions in the microscopic pictures which included

prepreg part and RFI part and had the co-cured interface area in the middle, and the widths of prepreg part and RFI part were both 1.0 ± 0.1 mm.

In addition, we selected five points which distributed uniformly on the four edges and center of specimens, and obtained the average thickness. Then, considering the void content, we calculated the fiber volume fractions based on the laminate thickness and fiber areal weight.¹²

The crack surface of samples after interlaminar fracture toughness testing was observed using scanning electron microscopy (SEM), Apollo 300 Field Emission, and the interfacial adhesion between fiber and resin was analyzed.

Results and discussion

Processing quality of co-curing process

The processing qualities of cured laminates using different processes were characterized by laminate thickness and optical micrograph.

As shown in Table 1, the thicknesses of laminates for different processes are similar. The thickness of co-cured laminates are slightly larger than those of laminates cured by RFI process and prepreg process. Compared with Process A, the isothermal dwell at 130°C applied in Process B has negligible influence on the thickness of laminates. In addition, the tackifier applied in the laminates using Process C leads to resin redundant, causing slightly increasing thickness.

Accordingly, the fiber volume fractions of the laminates are shown in Table 2. The fiber volume fractions of the prepreg laminate and the RFI laminate cured by Process B give the maximum values (about 60%), and the minimum from the co-cured laminate cured by Process C is about 57%. The range of the fiber volume fraction is from 56.9% to 60%, indicating that the fiber compaction of different laminates is slightly different.

The void contents of the laminates and the co-cured interfaces are listed in Table 3 and Table 4, respectively, where the corresponding standard deviations of some void contents ($\leq 0.03\%$) are too small ($< 0.01\%$) to be listed. As shown in Table 3, the quality of prepreg laminate is perfect without void and the prepreg/RFI laminate cured by Process A gives the maximum value (about 0.86%). In addition, the porosity volume fraction in the co-cured interface was also studied (shown in Table 4). The void contents in the co-cured interface are higher than that of the integral laminates, especially for prepreg/RFI laminates. The prepreg/RFI laminates have a higher porosity volume fraction than the RFI/prepreg laminates. It is attributed to the lay-up type which has important effects on the gas permeation through the laminates.

The micrographs of cross-sections of the RFI laminates fabricated by different processes are shown in Figure 7. Compared with the prepreg laminate with few void defects and resin-rich regions (see Figure 8), some matrix-rich regions and voids are found inside the RFI laminate cured by Process A presented in Figure 7(a). This is mainly ascribed to high viscosity of the resin film as presented in Figure 9.

As presented in Figure 9(a), the lowest viscosity of the resin film is approximately 10 Pa·s when the temperature is 130°C. The viscosity is high for resin infusion process.^{13,14} According to the isothermal viscosity-time curves (see Figure 9(b)), the viscosity at 130°C is less than 10 Pa·s for 1600 s, so we chose 130°C as the temperature point of the isothermal dwell applied to the cure cycle. Compared with the laminates cured by Process A (see in Figure 7(a)), the isothermal dwell applied in Process B has a great influence on the laminate resulting in an obvious decrease of voids (see in Figure 7(b)). This result is mainly attributed to the isothermal dwell (about 10 Pa·s at 130°C for 30 min) which ensures enough low resin viscosity and infusion time for improving the impregnation degree of the resin on the fiber preform.¹⁵ However, the isothermal dwell at 130°C has no obvious effect on the resin-rich region which is mainly caused by the fabric structure of a large tow and stitch-bonding line.

The micrographs of cross-sections of the co-RFI laminates fabricated by different processes are presented in Figures 10 and 11.

As presented in Figures 10 and 11, the resin-rich regions mostly exist in RFI part and the interface between the prepreg part and the RFI part. Moreover, there is an obvious difference in processing quality between prepreg/RFI laminates and RFI/prepreg laminates, i.e. some voids are found inside the prepreg/RFI laminates (see Figure 11), but few void defect forms inside the RFI/prepreg laminates (see Figure 10). Thus, the lay-up type has important effects

Table 1. Thickness of different laminates.

	Thickness (mm)		
	Process A	Process B	Process C
RFI laminate	3.97 ± 0.02	3.91 ± 0.04	4.08 ± 0.03
RFI/prepreg laminate	4.04 ± 0.02	4.02 ± 0.05	4.15 ± 0.04
Prepreg/RFI laminate	4.11 ± 0.02	4.09 ± 0.03	4.17 ± 0.04
Prepreg laminate	3.99 ± 0.02	—	—

RFI: resin film infusion.

Table 2. Fiber volume fraction of different laminates.

	Fiber volume fraction (%)		
	Process A	Process B	Process C
RFI laminate	59.1 ± 0.2	60.1 ± 0.3	57.5 ± 0.2
RFI/prepreg laminate	58.7 ± 0.1	59.0 ± 0.4	57.1 ± 0.3
Prepreg/RFI laminate	57.7 ± 0.3	58.0 ± 0.2	56.9 ± 0.1
Prepreg laminate	60.1 ± 0.2	—	—

RFI: resin film infusion.

Table 3. The void content of different laminates.

	Void content (%)		
	Process A	Process B	Process C
RFI laminate	0.03	0.02	0.01
RFI/prepreg laminate	0.03	0.01	0.01
Prepreg/RFI laminate	0.86 ± 0.03	0.63 ± 0.02	0.59 ± 0.01
Prepreg laminate	0	—	—

RFI: resin film infusion.

Table 4. The porosity volume fraction in the co-cured interface of the co-cured laminates.

	Porosity volume fraction (%)		
	Process A	Process B	Process C
RFI/prepreg laminate	0.03	0.02	0.02
Prepreg/RFI laminate	0.94 ± 0.03	0.76 ± 0.02	0.72 ± 0.01

RFI: resin film infusion.

on the gas permeation through the laminates and the void defects inside the laminates cured by co-RFI process.

As presented in Figures 7(c), 10(c) and 11(c), the qualities of laminates cured by Process C are similar to those of laminates fabricated by Process B, indicating that the tackifier has few effects on the laminates.

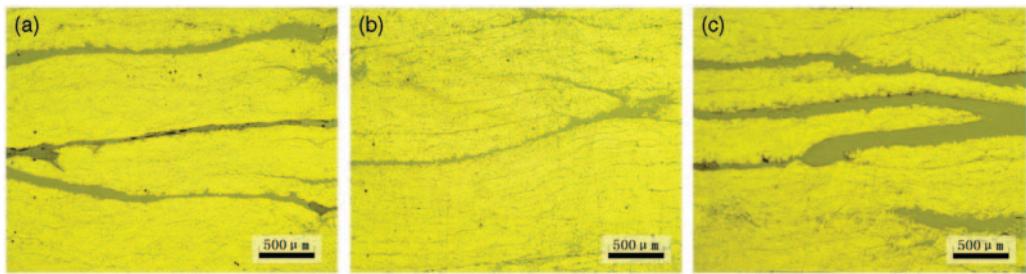


Figure 7. Micrographs of RFI laminates cured by: (a) Process A, (b) Process B and (c) Process C.
RFI: resin film infusion.

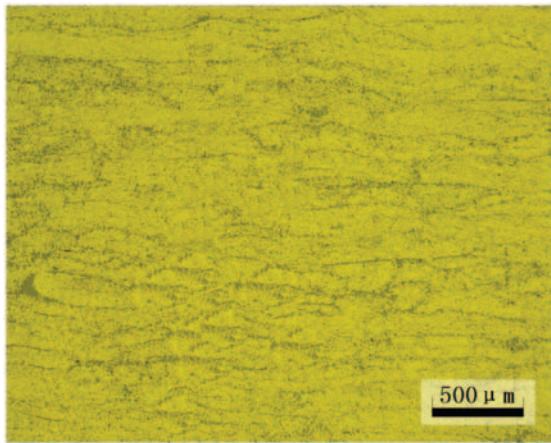


Figure 8. Micrographs of laminates cured by prepreg process.

The viscosity–temperature curve of the epoxy tackifier is presented in Figure 12, wherein the lowest viscosity is about 5 Pa·s when the temperature is ranged from 130°C to 230°C. Thus, the addition of the tackifier almost has no influence on the flow and infiltration of the matrix resin, but makes the resin redundant in comparison with the laminates fabricated without the tackifier.

Evaluation of co-RFI laminate interface

It is considered that the laminar interface which consists of prepreg part and RFI part is the unique characteristic for the co-curing structure. In this study, mode I and mode II interlaminar fracture toughness were adopted to evaluate the interlaminar properties of co-cured interface, and the specimens were cut from the laminates cured by Process A and prepreg-autoclave process.

Mode I interlaminar fracture toughness for initiation is often referred to as a standard material property of composite materials, which relates to a matrix-rich region ahead of the starter film being independent of fiber bridging effects.¹⁶ As shown in Figure 13, the

prepreg laminate has the maximum critical G_{IC} value (222 J/m^2) and the RFI processed laminate gives the minimum (168 J/m^2). The critical G_{IC} values of co-RFI laminates lie between the critical G_{IC} value of laminate manufactured by RFI process and that of laminate cured by prepreg-autoclave process. The critical G_{IC} values of RFI/prepreg laminate is 192 J/m^2 and that of prepreg/RFI is 190 J/m^2 .

Furthermore, mode I interlaminar fracture toughness for propagation was used to measure delamination resistance when the effect of fiber bridging and deflection of crack are involved, which require a higher driving force and cause a larger fracture area, and thus a higher fracture toughness.^{17–19} With the expansion of the delamination, there is a clear difference in the crack propagation pattern among the different composite laminates. The cracks in the prepreg laminate ran along the centerline without deflection. However, the cracks in other laminates propagated in meandering paths and at times through a different layer than initial crack layer, so the G_{IC} values of laminates cured by RFI and co-RFI process increase constantly and exceed that of prepreg laminate as presented in Figure 14. For the prepreg laminate, a better laminate quality with few matrix-rich regions and no void result in the maximum critical G_{IC} value. For the RFI laminate, the matrix-rich regions and voids (about 0.03%) reduce the critical G_{IC} value but the fiber bridging and the deflection of crack cause a higher fracture toughness at crack propagation stage. For the co-RFI laminates with more matrix-rich regions (see Figures 10(a) and 11(a)) and voids (see Table 4), the qualities of laminates are influenced by both the prepreg part and the RFI part, which cause the G_{IC} values of co-RFI laminates to lie between the G_{IC} value of RFI laminate and that of prepreg laminate. In addition, the void defects at the co-cured interface also reduce the interlaminar properties of the co-RFI laminates, causing a lower delamination resistance.

Figure 15 shows the SEM fracture morphology at delamination area of G_{IC} specimens. From Figure 16(a), little resin is adhered to the fiber surface of

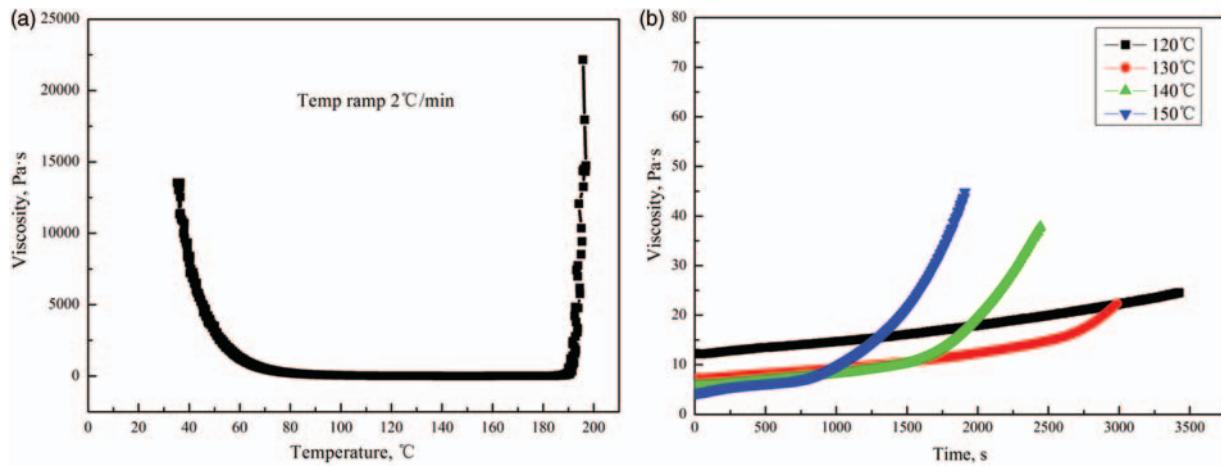


Figure 9. (a) Viscosity-temperature curve of resin film and (b) isothermal viscosity-time curves of resin film.

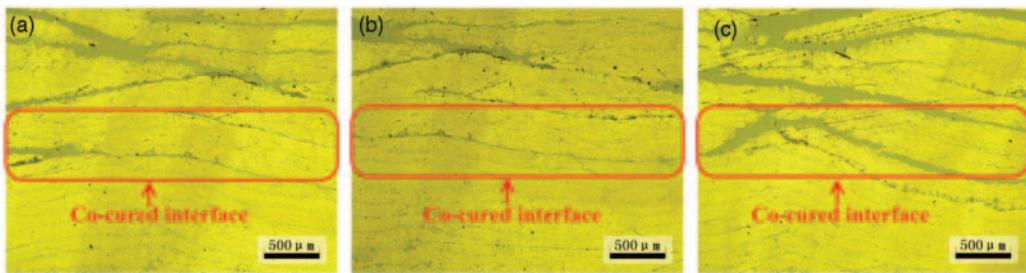


Figure 10. Micrographs of RFI/prepreg laminates cured by: (a) Process A, (b) Process B and (c) Process C. RFI: resin film infusion.

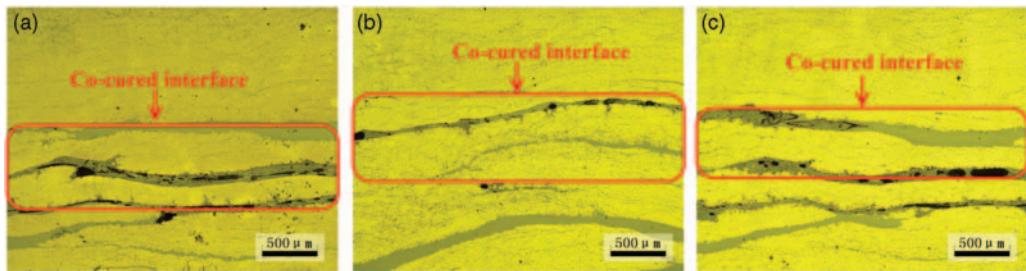


Figure 11. Micrographs of prepreg/RFI laminates cured by: (a) Process A, (b) Process B and (c) Process C.

prepreg laminates. In contrast, it is found that much resin remains on the fiber surface of the RFI laminates and the fracture surface is rough (see Figure 15(b)), indicating that the RFI laminates has a stronger interfacial adhesion than that of prepreg laminate. In addition, the lay-up type has little influence on the fracture surfaces of laminates cured by co-RFI procedure. The fiber surface of the prepreg part is rougher than that of prepreg laminate. On the other hand, for the morphology of RFI part, the fibers are coated with the resin which exhibits hackle markings as the result of crack extension. The different interfacial bonding between

prepreg laminate and RFI laminate results from the different surface characteristics of fibers in fabric and prepreg. Notice that the fiber varieties in CFW-200 fabric and MTM 44-1 prepreg are different. The different interfacial bonding cause different fiber bridging and deflection of crack, as shown in Figure 14.

Moreover, the interlaminar properties were also evaluated by ENF test for the laminates. Figure 16 presents the initial propagation G_{IIC} value of the composites laminates cured by Process A.

As is shown in Figure 16, the initial G_{Ic} value of prepreg laminate (about 2763 J/m²) is superior to that

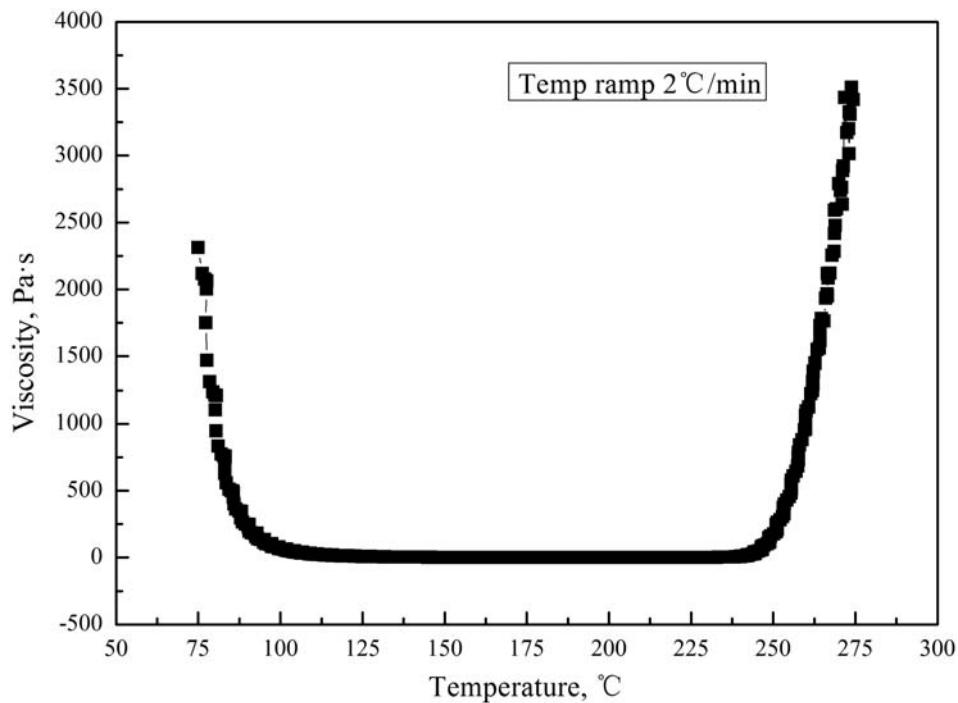


Figure 12. The viscosity–temperature curve of epoxy tackifier.

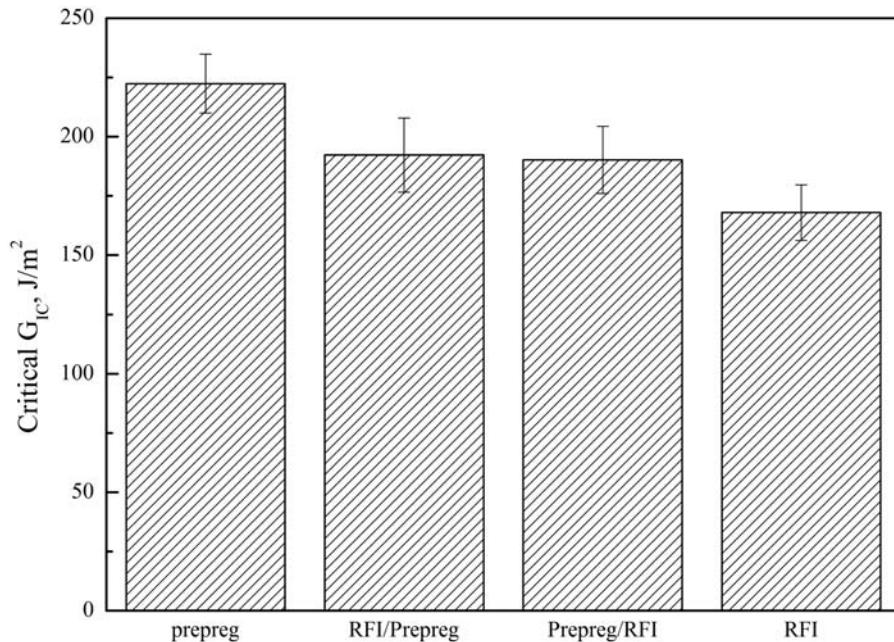


Figure 13. The critical G_{IC} values of different laminates fabricated using Process A.

of the laminate using RFI process which is approximately 548 J/m^2 . The initial G_{IIC} values of co-RFI laminates lie between the initial G_{IIC} value of laminate manufactured by RFI process and that of prepreg laminate. The initial G_{IIC} value of RFI/prepreg

laminate is 1415 J/m^2 and that of prepreg/RFI is 1155 J/m^2 . This result is mainly attributed to the matrix-rich regions and void defects formed inside the laminates.¹⁴ As presented in Figure 8 and Table 3, the prepreg laminate has excellent quality without

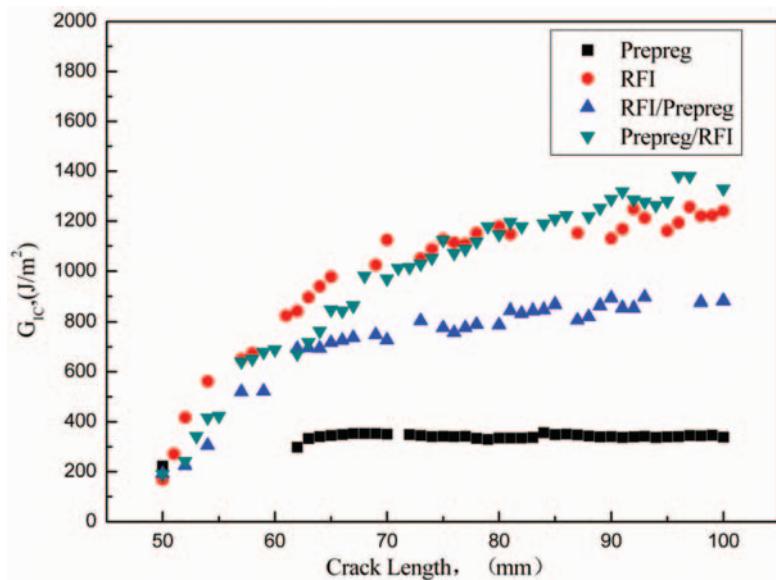


Figure 14. Mode I delamination resistance curves (R-curves) of laminates cured by Process A.

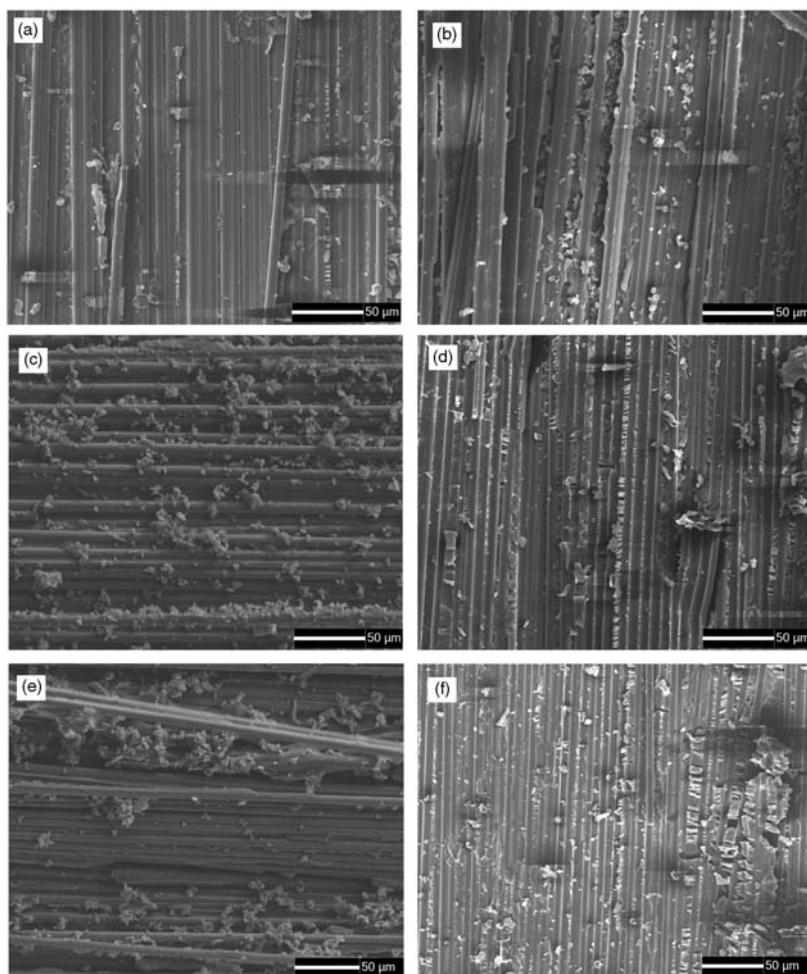


Figure 15. Fracture morphology at delamination area of G_{IC} samples: (a) prepreg laminate, (b) RFI laminate, (c) prepreg layer of RFI/Prepreg laminate, (d) RFI layer of RFI/Prepreg laminate, (e) prepreg layer of Prepreg/RFI laminate and (f) RFI layer of Prepreg/RFI laminate. All samples were fabricated using Process A.

RFI: resin film infusion.

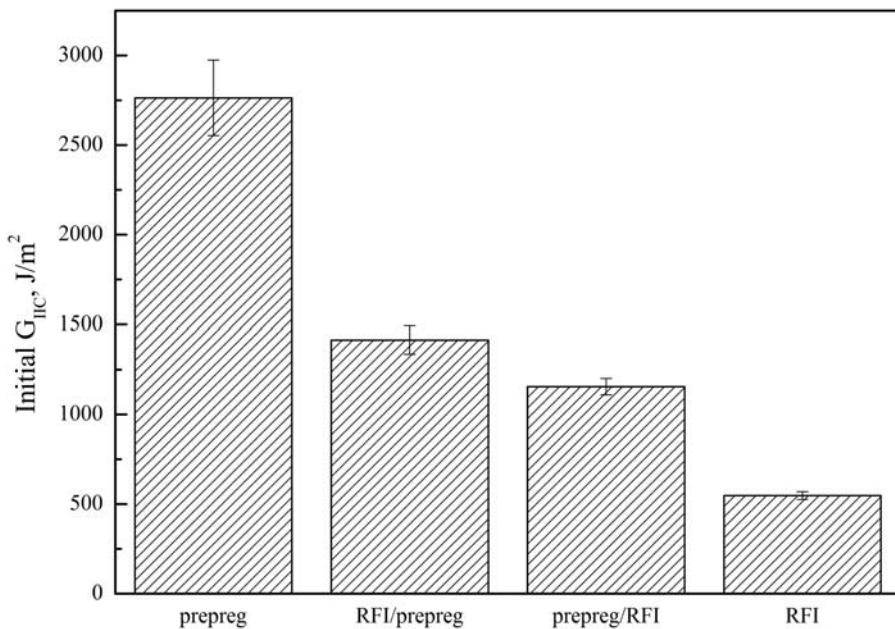


Figure 16. The initial G_{IC} values of different laminates cured by Process A.

void defect and matrix-rich region, while the RFI laminate and co-RFI laminates have poorer quality with more voids and matrix-rich regions, resulting in lower delamination resistance.

Influence of isothermal dwell and tackifier on the interlaminar properties

For RFI process, the isothermal dwell at 130°C was performed in Process B to ensure low enough resin viscosity and infusion time for improving the impregnation degree of the fiber preform.²⁰ In addition, epoxy tackifier was used to make fiber preform in Process C. Tackifier is usually adopted in practice for obtaining the initial dimensions and fiber distribution of preform before resin impregnation and curing process. Because the isothermal dwell and the tackifier influence the processing quality of co-RFI laminate (as mentioned in section ‘Processing quality of co-curing process’), the effects of the processing conditions on the mechanical properties should be considered. In this experiment, the laminates co-cured under Processes B and C were tested through the mode I and mode II interlaminar fracture toughness, and the effects of the isothermal dwell and the tackifier on the interlaminar properties of co-cured interface were evaluated.

Influence of isothermal dwell. The initial G_{IC} values of co-RFI laminates fabricated using Process A and Process B and the corresponding delamination resistance curves are presented in Figure 17 and Figure 18, respectively.

Figure 17 shows the average initial G_{IC} values of laminates cured by Process B and Process A. Standard deviation is also shown in Figure 17. The results show a significant increase in the initial G_{IC} values for the composite laminates cured by Process B, compared with those of laminates cured by Process A.

The samples were broken to inspect the surface of crack propagation, as shown in Figure 18. Compared to the surface of crack propagation of laminates cured by Process A (see Figure 15), more resin in the composites laminates cured by Process B is bonded to the carbon fibers. In addition, it is found that the fibers are coated with the resin which exhibits hackle markings as the result of crack extension from the morphology of RFI laminate, suggesting strong interfacial adhesion and crack propagation through the matrix in the composite laminates.²¹

Further studies on the interface of laminates cured by Process B have been taken with ENF. As shown in Figure 19, the isothermal dwell applied in the cure cycle improves the initial G_{IC} values and the increases of pre-peg/RFI co-curing laminate and RFI laminate are obvious.

Thus, both G_{IC} and G_{HIC} of the composite laminates cured by Process B are higher than those of the laminates cured by Process A, indicating that the isothermal dwell has a positive influence on the interlaminar fracture toughness of laminates. The isothermal dwell makes resin flow more fully and reduces the matrix-rich regions and void defects formed in the laminates, resulting in the better interlaminar properties.

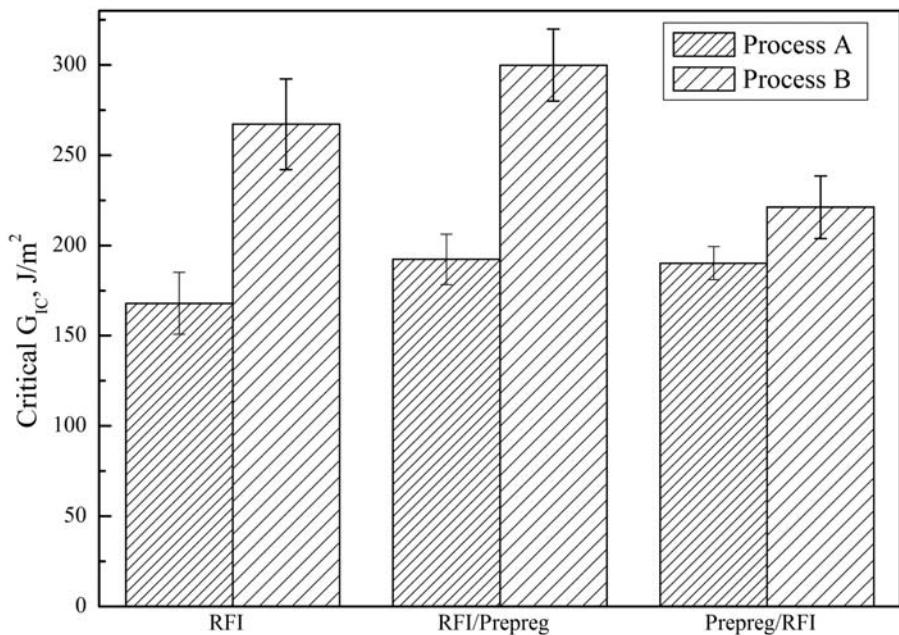


Figure 17. The initial G_{IC} values of laminates cured by Process A and Process B.

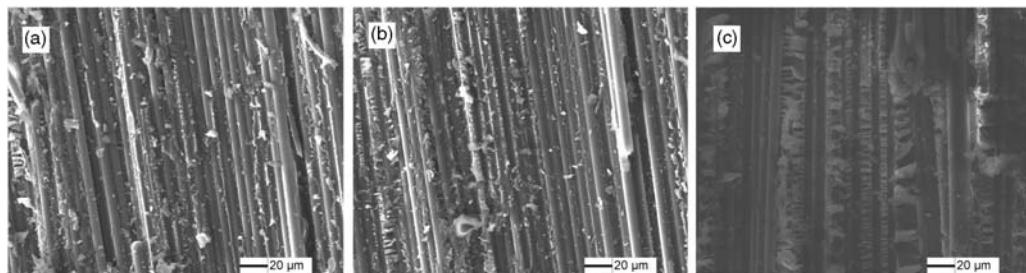


Figure 18. Fracture morphology at delaminate area of G_{IC} samples cured by Process B: (a) prepreg layer of co-RFI laminate, (b) RFI layer of co-RFI laminate and (c) RFI laminate.

RFI: resin film infusion.

Influence of tackifier. The critical G_{IC} values of composite laminates fabricated using Process B and Process C are presented in Figure 20.

As shown in Figure 21, the tackifier has effect on the interlaminar fracture toughness of laminates. The initial G_{IC} value of RFI laminate with tackifier is improved compared with the RFI laminate without tackifier, but the initial G_{IC} values of co-RFI laminates with tackifier decrease in contrast with those of co-RFI laminates without tackifier. That is mainly attributed to two factors. On the one hand, epoxy tackifier can enhance interlaminar fracture toughness with the addition of the toughened resin particles,²² but on the other hand, the tackifier makes the resin redundant and enlarges the matrix-rich region which can cause the reduction of the initial G_{IC} values.

The surfaces of crack propagation of the samples were inspected with SEM, as shown in Figure 21. In comparison with the surface of crack propagation of the laminates cured by Process B (see Figure 18), the crack surfaces of the laminates cured by Process C with tackifier present few differences, which proves the tackifier has few influence on the impregnation of the matrix resin on fiber.

The interlaminar properties of the laminates with tackifier were also evaluated by ENF test. The initial propagation G_{IIIC} values are presented in Figure 22. It is obvious that the tackifier has a great effect on the mode II interlaminar fracture toughness. Compared with the laminates without tackifier, the addition of the tackifier improves the initial G_{IC} values of the RFI laminate and the RFI/prepreg laminate, but the initial G_{IC} value of

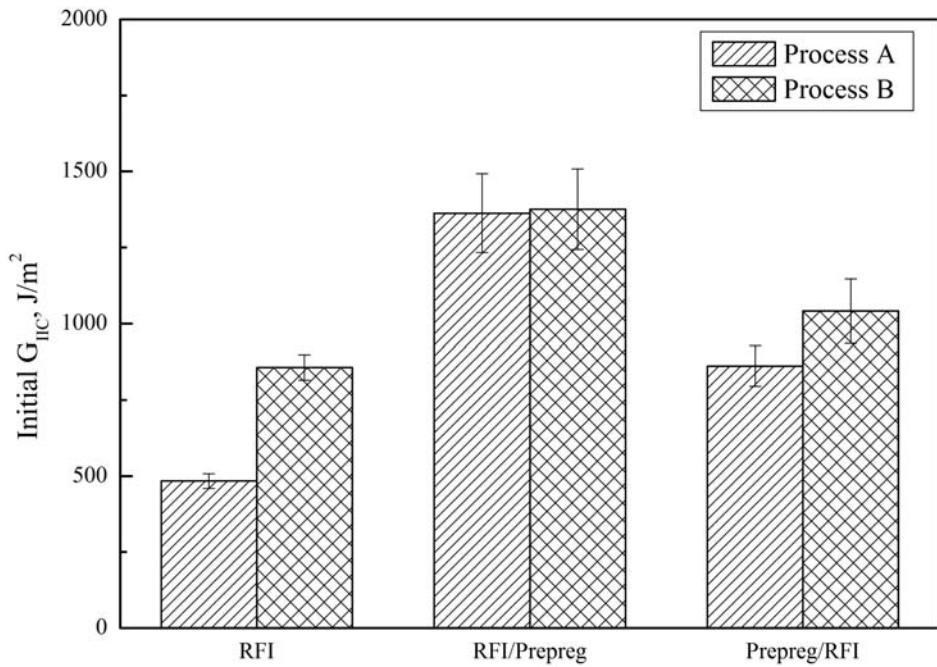


Figure 19. The initial G_{IIC} values of laminates cured by Process A and Process B.

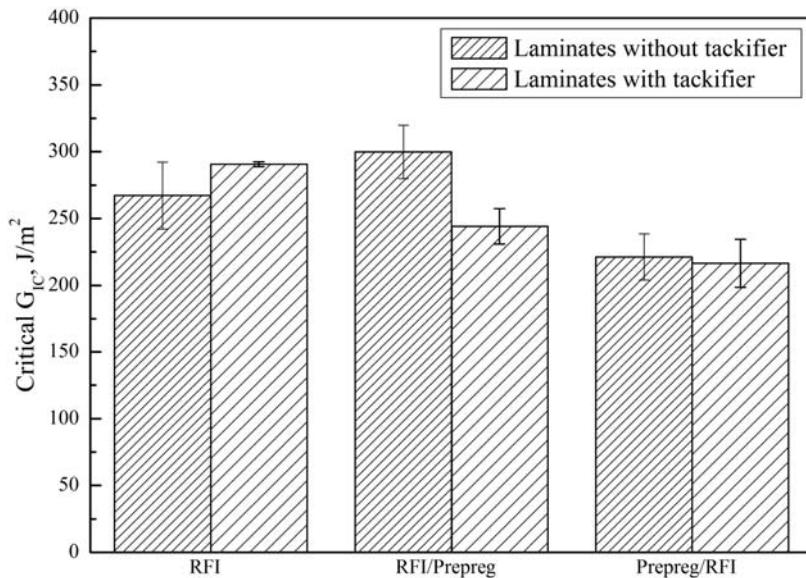


Figure 20. The critical G_{IC} values of laminates cured by Process B and Process C.

the prepreg/RFI laminate decreases because of the expansion of matrix-rich region caused by the addition of the tackifier (Figure 11).

In addition, the results including the initial G_{IC} values and the initial propagation G_{IIC} values of different laminates fabricated using Processes A, B and C are presented in Table 5 and Table 6, respectively. The RFI/prepreg laminates with the same processes have larger initial G_{IC} values and initial G_{IIC} values than

the prepreg/RFI laminates. The RFI/prepreg laminates cured by Processes A and B have better interface properties in comparison with that of RFI laminates. Furthermore, both the initial G_{IC} values and the initial G_{IIC} values of the laminates cured by Process B have improved in different degree in comparison with those of the laminates cured by Process A. It indicates that the weakness of the co-cured interface area can be eliminated, and the process design has important effects

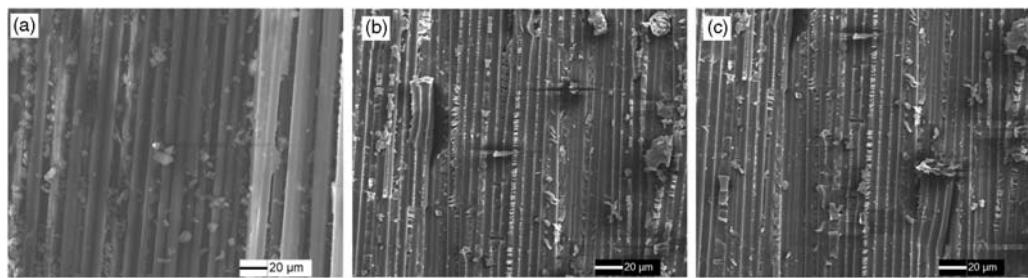


Figure 21. Fracture morphology at delamination area of laminates with tackifier: (a) prepreg layer of co-RFI laminate, (b) RFI layer of co-RFI laminate and (c) RFI laminate.

RFI: resin film infusion.

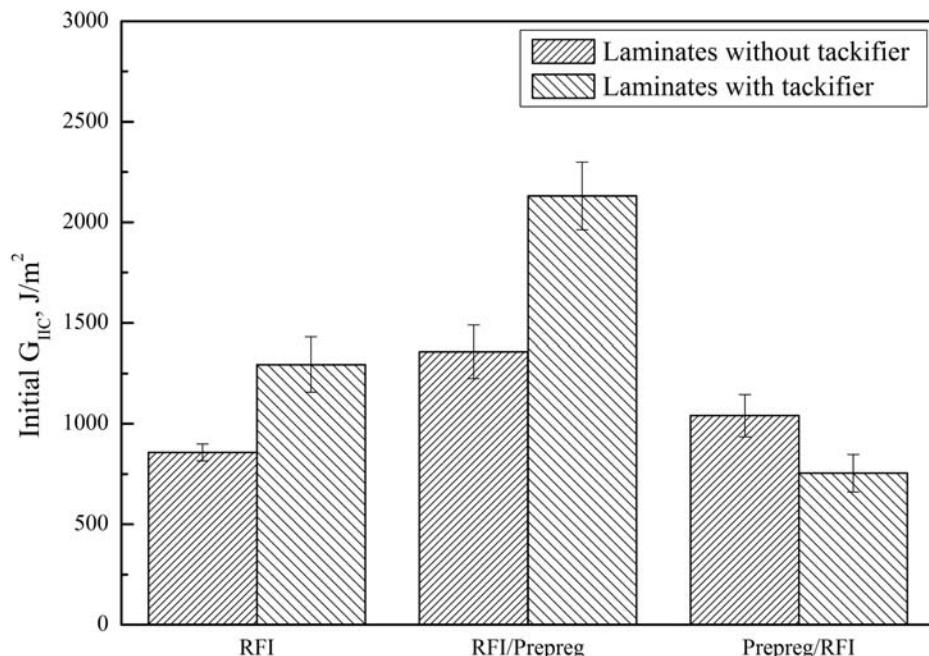


Figure 22. The initial G_{IC} values of different laminates with and without tackifier.

Table 5. The initial G_{IC} values of laminates cured by Processes A, B and C.

	The initial G_{IC} values		
	Process A	Process B	Process C
RFI laminate	167.96 ± 17.09	267.17 ± 25.09	290.59 ± 1.84
RFI/prepreg laminate	192.28 ± 14.01	299.87 ± 19.95	244.15 ± 13.21
Prepreg/RFI laminate	190.18 ± 9.10	221.17 ± 17.35	216.44 ± 17.93

RFI: resin film infusion.

on the processing quality and the co-cured interface properties.

Thus, in order to obtain co-RFI laminates with excellent processing quality and interface properties, a

Table 6. The initial G_{IIIC} values of laminates cured by Process A, B and C.

	The initial G_{IIIC} values		
	Process A	Process B	Process C
RFI laminate	547.96 ± 21.58	1159.90 ± 53.34	1838.77 ± 72.05
RFI/prepreg laminate	1414.77 ± 78.15	2031.17 ± 135.70	2553.02 ± 109.44
Prepreg/RFI laminate	1155.205 ± 46.37	1776.62 ± 169.335	984.44 ± 126.98

RFI: resin film infusion.

suitable cure cycle is necessary to ensure enough low resin viscosity and infusion time for improving the impregnation degrees of the fiber preform and the co-cured interface. In addition, the effects of tackifier

on resin flowing and interlaminar resin-rich region should be taken into account, which can change the interlaminar fracture toughness at the co-cured interface.

Conclusion

This article presents a co-curing composite molding process combining RFI with prepreg, named co-RFI, and the carbon fiber/epoxy resin matrix composite laminates were fabricated using the developing co-curing process with unidirectional prepreg and its corresponding resin film.

Optical photographs of the laminates cured by the co-RFI process show that the matrix-rich regions and voids mostly exist in RFI part and the interlaminar interface between the prepreg part and RFI part. The matrix-rich regions and voids at the co-cured interface reduce the interlaminar properties of the co-RFI laminates, causing a lower delamination resistance. The lay-up type of prepreg part and RFI part has a great influence on the quality of the composite laminates, where locating prepreg part below RFI part is favorable for eliminating voids in laminates. In comparison with the laminates manufactured by RFI process and prepreg-autoclave process, the critical interlaminar fracture toughness at the co-cured interface of the laminates cured by the co-curing process lies between the prepreg laminates and the RFI laminates, and G_{IC} at crack propagation stage for co-RFI laminate is higher due to fiber bridging and deflection of crack. Furthermore, the isothermal dwell before curing process has a great influence on the interlaminar fracture toughness of co-RFI laminates, which improves the interlaminar fracture toughness, resulted from enough low resin viscosity and infusion time for improving the impregnation degree of the fiber preform. In addition, the epoxy tackifier is also an important factor, which makes the resin redundant and enlarges the matrix-rich region at the co-cured interface, and changes the interlaminar fracture toughness. These results have close relationships with the compacting structure of fibers in prepreg and RFI parts and the interfacial bonding between the two kinds of fiber and the matrix.

Funding

This work was financially supported by the Fund of National Engineering and Research Center for Commercial Aircraft Manufacturing (Project No. SAMC12-JS-15-010) and the National 973 Program of China (Project No. 2010CB631100).

Conflict of interest

None declared.

References

- Qi B, Raju J, Kruckenberg T, et al. A resin film infusion process for manufacture of advanced composite structures. *Compos Struct* 1999; 47(1–4): 471–476.
- Garschke C, Weimer C, Parlevliet PP, et al. Out-of-autoclave cure cycle study of a resin film infusion process using in situ process monitoring. *Compos Part A* 2012; 43(6): 935–944.
- Antonucci V, Giordano M, Nicolais L, et al. Resin flow monitoring in resin film infusion process. *J Mater Process Tech* 2003; 143–144: 687–692.
- Marguerès P, Périé J, Perez JG, et al. Characterization of a composite structure obtained by RFI using HexFIT® semi-products. *Compos Sci Technol* 2009; 69(1): 117–124.
- Sheu CH, Shimazu DM and Kane DM. *Co-cured vacuum-assisted resin transfer molding manufacturing method*. US Patent: US 2004/0051214 A1[P], 2004.
- Husmann CH, Sheu CH and Shimazu DM. *Co-cured resin transfer molding manufacturing method*. US Patent: US 7374715 B2[P], 2008.
- Kaps R. Combined prepreg- and infusion technology for integral composite structures. *DLR Dtsch Zent Luft-Raumfahrt Forschungsber* 2011; 34: 1–179.
- Kaps R, Herbeck L and Herrmann A. Hybrid fabrication route-cost efficient CFRP primary airframe structures. In: *ICAS 2006, 25th international congress of the aeronautical sciences*, Hamburg, Germany, 3–8 September 2006.
- Thakre PR, Lagoudas DC and Riddick JC. Investigation of the effect of single wall carbon nanotubes on interlaminar fracture toughness of woven carbon fiber-epoxy composites. *J Compos Mater* 2011; 45(10): 1091–1107.
- Shokrieh MM, Heidari-Rarani M and Ayatollahi MR. Interlaminar fracture toughness of unidirectional DCB specimens: a novel theoretical approach. *Polym Test* 2012; 31(1): 68–75.
- Davis DC and Whelan BD. An experimental study of interlaminar shear fracture toughness of a nanotube reinforced composite. *Compos Part B* 2011; 42(1): 105–116.
- Xin CB, Gu YZ, Li M, et al. Online monitoring and analysis of resin pressure inside composite laminate during zero-bleeding autoclave process. *Polym Composite* 2011; 32(2): 314–323.
- Liang GZ and Wang D. High-performance bismaleimide resin for resin film infusion. *Polym-Plast Technol* 2002; 41(2): 195–285.
- Liang B, Chen LX, Dong JN, et al. A thermosetting phenolic film and its application in the resin film infusion. *J Compos Mater* 2011; 46(3): 190–285.
- Celle P, Drapier S and Bergheau J. Numerical modelling of liquid infusion into fibrous media undergoing compaction. *Eur J Mech A Solids* 2008; 27(4): 647–661.
- Zhang D, Ye L, Deng S, et al. CF/EP composite laminates with carbon black and copper chloride for improved electrical conductivity and interlaminar fracture toughness. *Compos Sci Technol* 2012; 72(3): 412–420.
- Yokozeki T, Iwahori Y and Ishiwata S. Mechanical properties of CFRP laminates manufactured from

- unidirectional prepgs using CSCNT-dispersed epoxy. *Compos Part A* 2007; 38(10): 2121–2130.
- 18. Siddiqui NA, Woo RSC and Kim JK. Mode I interlaminar fracture behavior and mechanical properties of CFRPs with nanoclay-filled epoxy matrix. *Compos Part A* 2007; 38(2): 449–460.
 - 19. Mathews MJ and Swanson SR. Characterization of the interlaminar fracture toughness of a laminated carbon/epoxy composite. *Compos Sci Technol* 2007; 67(7–8): 1489–1498.
 - 20. Davies LW, Day RJ, Bond D, et al. Effect of cure cycle heat transfer rates on the physical and mechanical properties of an epoxy matrix composite. *Compos Sci Technol* 2007; 67(9): 1892–1899.
 - 21. Rikards R, Korjakin A, Buchholz FG, et al. Interlaminar fracture toughness of GFRP influenced by fiber surface treatment. *J Compos Mater* 1998; 17(32): 1528–1559.
 - 22. Kusaka T, Watanabe K, Hojo M, et al. Fracture behavior and toughening mechanism in Zanchor reinforced composites under mode II loading. *Compos Sci Technol* 2009; 69(14): 2323–2330.