International Journal of Adhesion & Adhesives 19 (1999) 231–242

Improved 121°C curing epoxy film adhesive for composite bonding and repair applications: FM® 300-2 adhesive system

Dalip K. Kohli

CYTEC-Fiberite Inc., Havre de Grace, Maryland, USA

Abstract

Performance of a high Tg, dual 121°C/177°C curing epoxy adhesive film FM® 300-2 is described for bonding of both thermoplastics and thermoset composite substrates. Effects of humidity, laminate layup and composite surface preparation on the durability of the bonded structure are discussed. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Adhesive; Aerospace; Metal bonding; Composite bonding; Repair

1. Introduction

Adhesive bonding is the most suitable method of joining both metallic and non-metallic structures where strength, stiffness and fatigue life must be maximized at a minimum weight. Use of tough, moisture resistant, flow controlled, epoxy based adhesives to bond composites to composites, or composites to metals offers a number of advantages [1]. Because of their high elongation and high ultimate shear strength, these adhesives are particularly suitable for redistributing the high shear stress concentrations of composite metal bonds and in accommodating the low interlaminar shear strength of the composite.

For aerospace bonding applications, FM® 300 epoxy based film adhesive is widely used for bonding metal to composite structures. Some of these applications range from bonding the wing-root assemblies (titanium to graphite epoxy), to composite sandwich structures on F-18 fighter aircraft, to bonding and surfacing applications on most commercial and military aircraft worldwide. This 177°C curing adhesive is used in both co-cure and secondary bonding applications.

In the case of bonding metal to composite parts, the 177°C cure temperature for secondary bonding can lead to significant thermal stresses due to the difference in coefficient of thermal expansion between the metal and the composite. These induced stresses can result in the loss of dimensional stability, disbonds, or delamination in these parts. In order to overcome these problems, there is a need for a durable 121°C curing film adhesive that provides the performance of 177°C curing film adhesives in both metal and composite bonding applications.

Recently, we have reported on the development of such an adhesive system for aerospace bonding applications. This adhesive film designated FM® 300-2 is a 121°C cure version of 177°C curing FM® 300 adhesive film and provides similar stress-strain and mechanical performance to the FM® 300 system [2, 3].

This paper describes the performance of this 121°C curing, high Tg film adhesive for composite bonding and repair applications.

2. Materials and procedures

2.1. Adhesive cure cycle

For all metal or secondary bonding of composite substrates, the following cure cycle was utilized: Heat-up rate 1.7°C/min to 121°C, 0.28 mpa, hold 90 min at 121°C.

2.2. Secondary bonding of composites

2.2.1. Thermosetting composites

For secondary bonding work, eight epoxy/graphite or epoxy/glass prepreg systems with cure temperatures ranging from 121°C to 177°C were selected. The prepregs and their cure cycles are listed below.

CYCOM® 919/3K70P, epoxy graphite fabric prepreg – cured 60 min at 121°C, 0.35 mPa, laminate thickness 2.5 mm.

PII: S0143-7496(98)00038-4

CYCOM® 985/3K70P, epoxy/graphite fabric prepreg – cured 120 min at 177°C, 0.52 mPa, heat-up rate 1.7°C/min, laminate thickness 2.5 mm.
CYCOM® 1827/6681 epoxy/glass fabric prepreg – cured 120 min at 177°C, 0.52 mPa, heat-up rate 1.7°C/min, laminate thickness 2 mm.
Hercules 8551-7/AS-4, epoxy/graphite fiber tape – cured 120 min at 177°C, 0.35 mPa, heat-up rate 1.7°C/min, laminate thickness 1.27 mm, laminate lay-up $(\pm 45, \pm 90, 0)_2$.
Hercules 8551-7/glass, epoxy/glass fiber tape – cured 120 min at 177°C, 0.35 mPa, heat-up rate 1.7°C/min, laminate thickness 1.27 mm, laminate lay-up $(\pm 45, \pm 90, 0)_2$.
Hercules 3501-6/3K70P, epoxy/graphite fabric prepreg – cured 120 min at 177°C, 0.35 mPa, heat-up rate 1.7°C/min, laminate thickness 3.1 mm.
Ciba 6376/3K70P, epoxy/graphite fabric prepreg – cured 120 min at 177°C, 0.35 mPa, heat-up rate 2°C/min, laminate thickness 3.1 mm.
Ciba M20, a 121°C curing graphite/epoxy prepreg-cured 120 min at 121°C and 0.35 mPa, heat-up rate 1.5°C/min, laminate thickness 3.1 mm.

2.2.2. Thermoplastic composites

For secondary bonding of thermoplastic laminates APC-2 (PEEK) or KIII, laminates were used as received from the manufacturer.

2.2.3. Surface preparation

Types of surface preparation are listed with the tabulated data.

For Hercules 8551-7 precured graphite and glass laminates, no peel ply was used. Instead, the effect of three different surface preparations (a) solvent wipe only, (b) hand sand/solvent wipe, and (c) sandblast/solvent wipe was studied. For solvent wiping, methyl ethyl ketone was used.

For hand sanding, a 320 grit sand paper was used and for sandblasting, 150 to 200 grit Al_20_3 at 0.28 mPa (40 psi) pressure was used.

2.3. Co-cure studies

For co-cure work, CYCOM® 985/AS-4, a 177°C curing epoxy/graphite prepreg and Ciba M 20/3K70P, a curing epoxy/graphite prepreg were used. The adhesive was co-cured with the prepreg at either 177°C or 121°C. For surface ply studies, FM® 300-2M, 150 gm/m^2 adhesive film was used as the surface layer and co-cured with the prepreg.

2.4. Test methods

For mechanical testing the following test methods were used: Lap Shear – ASTM D1002, Floating Roller Peel – ASTM D3167, Honeycomb Sandwich Peel – ASTM D1781, Flatwise Tensile – ASTM C297.

3. Results and discussion

Utilizing an innovative materials approach, we have developed a 121°C cure version of FM® 300 adhesive for various bonding applications where higher performance is needed but the cure temperature can not be higher than 121°C.

This adhesive system, designated as FM® 300-2, is based on epoxy chemistry and is designed for bonding metallic and composite structures as well as structures fabricated from metallic, Nomex or fiberglass honeycomb. This adhesive system has been formulated to fully cure within 90 min at 121°C and has flow and handling properties similar to FM® 300 adhesive.

Comparison of physical and mechanical property data between these adhesives is shown in Table 1. It is notable that the 121°C curing adhesive has a glass transition temperature of 144°C as compared to 148°C for the 177°C cured. Because of this high glass transition temperature, this film adhesive is capable of significant retention of its strength up to 150°C.

The toughness properties of this adhesive as indicated by peel strength are very similar to FM® 300. Both these systems have metal to metal peel strength of 6.4 kN/m (35 pli). Because of their similar toughness and flow properties, both adhesives have similar sandwich properties.

One of the key objectives in the development of this new adhesive was that it should have stress-strain properties similar to the adhesive. The comparative

Table 1
FM® 300-2 adhesive film baseline comparison with FM® 300 adhesive film aluminum adherends

Property	FM 300-2K cured 90 min at 121°C	FM 300K cured 60 min at 177°C
Lap shear strength (mPa (psi))		
24°C	38.6 (5600)	37.9 (5500)
121°C	26.9 (3900)	27.6 (4000)
150°C	15.9 (2300)	18.6 (2700)
Floating roller peel (kN/m (pli))		
24°C	6.4 (36)	6.2 (35)
Honeycomb sandwich peel (N m/m (ipp3in)) 24°C	74 (50)	67 (45)
Flatwise tensile (mPa (psi))		
24°C	7.6 (1100)	6.9 (1000)
150°C	2.8 (400)	3.2 (400)
Flow (%)	450–550	450–550
Tg°C (TMA)	144	148

Note: Primer: BR® 127, 0.005 mm thick, cured 60 min at 121°C; Metal: 2024-T3 FPL etched. Adhesive weight: 392 gm/m^2.

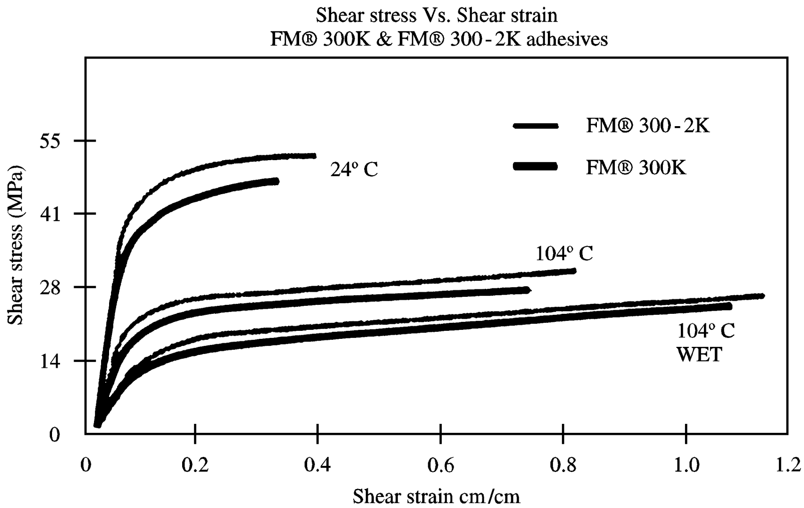

Fig. 1.

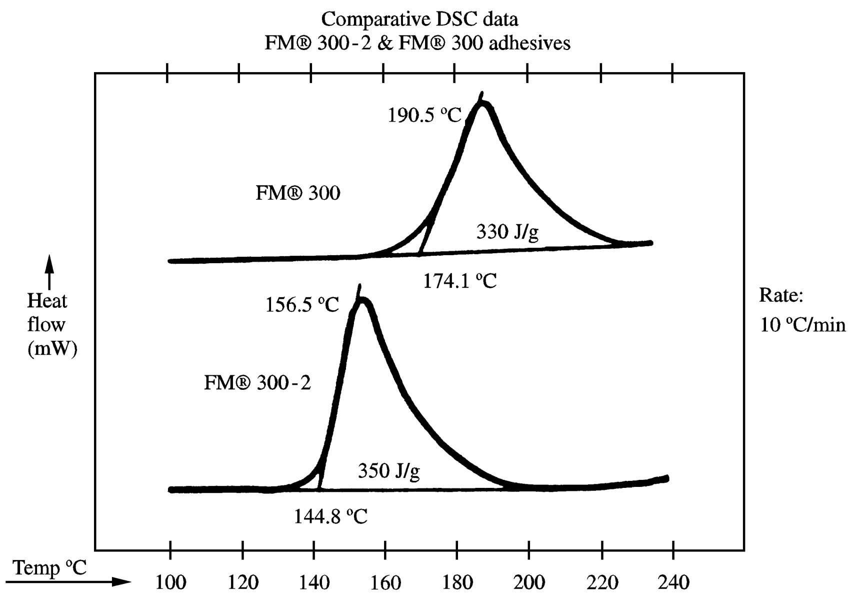

Fig. 2.

stress-strain properties of these two adhesives are shown in Fig. 1. Note that the 121°C cured FM® 300-2 adhesive has the same stress and strain properties as FM® 300 adhesive, not only under dry but also under hot/wet conditions up to 104°C. This means that the lower temperature curing adhesive can be used to minimize stresses caused by differential coefficients of expansion when bonding dissimilar substrates.

The DSC profiles for these adhesives are shown in Fig. 2. The onset temperature for FM® 300-2 adhesive is approximately 121°C as compared to 150°C for the FM® 300 system. These onset temperatures indicate the different cure chemistry operating in these two systems.

All the comparative data generated thus far indicates that this new adhesive provides similar stress-strain and mechanical properties to its higher temperature curing version. The comparative composite bonding data between these two adhesives is discussed later.

3.1. *Effect of cure cycle*

The ability of adhesives to cure over a wide temperature range becomes significant when these adhesives are to be used for co-cure and secondary bonding of 121°C to 177°C curing epoxy prepreg systems.

Since this new adhesive is designed to be used for these applications, we evaluated the effect of changing the cure temperature on its physical and mechanical properties. These data are summarized in Table 2. Examination of the data in Table 2 shows no discernible trends. Even the glass transition temperature does not significantly change with the increase in cure temperature from 121°C to 177°C. This implies that 121°C cure temperature is sufficient to complete the crosslinking in this adhesive system. We have also evaluated the effect of thermal cycling at 177°C on the lap shear and peel strength of this adhesive system. In this study, bonded coupons were exposed to 177°C for two to eight hours and then tested at various temperatures. Results from this study indicate that there is no drop off in the shear or peel strength after short-term thermal cycling at 177°C (Table 3).

Results from the cure cycle study indicate that this adhesive can be cured over a wide temperature range of 121°C to 177°C without change in its bonding performance.

3.2. *Effect of humidity*

The effect of prebond humidity was evaluated by exposing the adhesive film and the metal substrates to two weeks at 80% relative humidity at 24°C. For postbond humidity studies, individually cut coupons were exposed to 71°C and 100% relative humidity for 30 days. The data is shown in Table 4a. The FM® 300-2 adhesive shows excellent retention of its strength up to 121°C after prebond and postbond humidity exposures.

The outstanding moisture resistance of this adhesive is also demonstrated by its ability to bond to wet Nomex and maintain its strength up to 121°C even after humidity exposures. The wet Nomex honeycomb bonding data is shown in Table 4b.

Table 4a
Effect of prebond and postbond humidity exposures on peel strength. FM 300-2K adhesive film BR127 adhesive primer

Prebond film exposure	Bonded coupon exposure	Floating roller peel (kN/m (pli))	
		24°C	121°C
None	None	6.3 (36)	6.3 (36)
	30 days at 71°C/100% RH	6.2 (35)	6.9 (39)
15 days at 80% RH at 24°C	None	7.0 (40)	6.5 (37)
	30 days at 71°C/100% RH	5.6 (32)	6.3 (36)

Note: Adhesive weight: 392 gm/m^2.

Table 2
Effect of cure cycle on physical properties of FM® 300-2K (392 gms/m^2) 2024T3 aluminum adherends

Test	Test temp. (°C)	Cure cycle		
		90' at 121°C	60' at 150°C	60' at 177°C
Lap shear strength (MPa(psi))	24	42.0 (6100)	44.5 (6455)	43.2 (6280)
	107	32.1 (4660)	32.2 (4680)	31.4 (4560)
Floating roller peel (kN/m (pli))	24	5.3 (30)	6.0 (34)	6.0 (34)
	107	7.6 (43)	7.9 (45)	8.6 (49)
HCSP (Nm/m (in-lbs/3 in))	24	71 (48)	77 (52)	74 (50)
	107	74 (50)	81 (55)	60 (89)
Flow (%)		575	525	575
Tg (°C (TMA))		143	140	140

Note: Primer: BR® 127, 0.005 mm thick, cured at 121°C for 60 min.

Table 3
Effect of 177°C cycling on lap shear and floating roller peel strength of FM® 300-2K (392 gms/m^2) adhesive aluminum adherends

Bonded coupon exposure	LSS (MPa (psi))		Floating roller peel (kN/m (pli))	
	24°C	104°C	24°C	104°C
No additional exposure	43.9 (6370)	36.1 (5250)	4.9 (28)	7.0 (40)
Exposed to 1 cycle[a] at 177°C	49.3 (7160)	37.0 (5380)	4.2 (24)	6.0 (34)
Exposed to 2 cycles[a] at 177°C	47.1 (6840)	37.6 (5466)	4.6 (27)	6.3 (36)
Exposed to 3 cycles[a] at 177°C	47.7 (6925)	37.2 (5410)	4.6 (26)	6.3 (36)

[a]Cycle: Heat 24°C to 177°C, hold at 177°C for 2 h, cool to 24°C.
Note: *Adhesive cure cycle*: 90 min at 121°C, 0.27 mPa, 1.7°C/min heat up rate. Primer: BR® 127, 0.005 mm thick, cured 60 min at 121°C.

Table 4b
Wet Nomex bonding with FM 300-2K adhesive film

Nomex core exposure	Bonded coupon exposure	Flatwise tensile (mPa (psi))		
		24°C	82°C	120°C
	None	5.4 (780) (Core)	5.3 (775) (Core)	4.3 (625) (Core)
3 weeks at 65% RH at 24°C	30 days at 71°C 100% RH	4.8 (700) Core	3.9 (570) (Core)	— —
Immersed in water fot 24 h at 60°C then bonded within 2 h	None	5.0 (720) (Core)	5.1 (745) (Core)	4.2 (615) (Core)
	30 days at 71°C 100% RH	5.0 (725) (Core)	3.7 (545) (Core)	— — (Core)

Note: Failure mode: 100% Core. Adhesive weight: 392 gm/m^2.

4. Composite bonding

A major goal in the development of this adhesive film was that it should be suitable for use in co-cure applications with 121°C and 177°C cure prepregs as well as for bonding to the surfaces of properly prepared thermoset and thermoplastic laminates. One of the problems with the currently used 121°C curing adhesive films is that these are affected by the moisture in the precured laminate or the moisture in the non-metallic core. This moisture degrades the performance of these adhesive systems by plasticization as well as due to the porosity in the bondline.

As part of our studies, we wanted to investigate the performance of FM® 300-2 under a myriad of conditions to determine if the need for currently drying the Nomex core or the precured composite skins could be eliminated. Also, we needed to define the appropriate surface preparation for the precured substrates.

4.1. Effect of precured composite substrate exposures

The effect of precured composite substrate conditioning was studied by exposing the cured laminate to various conditions and then bonding the lap shear panels with the FM 300-2 adhesive film. The bonded specimens were then tested before and after humidity exposures. The wet conditioning of the bonded specimens involved exposure to 71°C/100% RH for 30 days. Test results are shown in Tables 5 and 6.

The results in Table 5, summarize the testing on the Ciba 6376 precured substrates. These results show the effect of predrying the laminate to completely saturating the laminate with humidity prior to bonding. Test data indicates that the performance of this adhesive system is not significantly affected by the exposure to moisture from the laminate during the cure. This is a remarkable result and shows that there is no need to dry the composite laminates prior to bonding with the FM 300-2 adhesive film.

Table 5
Secondary bonding of Ciba 6376/3K70 Prepreg, with FM® 300-2M. Effect of precured substrate conditioning on carbon–carbon lap shear strength

Property	Test temp.	Test results Prebond substrate conditioning					
		1 h dry @ 82°C	1 h dry @ 121°C	30 days @ 23°C	1 day @ 60°C/ 100% RH	5 days @ 60°C/ 100% RH	10 days @ 60°C/ 100% RH
Carbon lap shear		19.25	18.9	18.7	18.0	17.87	18.5
(mPa (psi))	− 55°C	(2790)	(2745)	(2715)	(2615)	(2590)	(2680)
Peel ply	23°C	20.7	20.3	18.6	23.1	24.7	22.5
Surface		(3000)	(2950)	(2695)	(3350)	(3580)	(3255)
Preparation	82°C/	27.9	29.0	26.1	5.8	16.3	16.3
	Wet[a]	(4050)	(4200)	(3790)	(2295)	(2365)	(2355)

[a]Wet conditioning: 30 days at 71°C/100% RH.
Adhesive weight: 300 gm/m^2.

Table 6
Secondary bonding of Hercules 3501-6/3K70P Prepreg with FM® 300-2 adhesive film effect of precured composite substrate conditioning on the carbon-carbon lap shear strength

Test temperature	Test results Prebond substrate conditioning		
	1 h @ 82°C	30 d at @ 23°C	10 d @ 60°C/ 100% RH
− 55°C	22.6 (3280)	21.6 (3130)	17.2 (2490)
23°C	28.7 (4160)	28.5 (4135)	26.2 (3795)
82°C/Wet[a]	30.2 (4380)	24.8 (3590)	14.6 (2120)

[a]Wet conditioning: 30 days at 71°C/100% RH.
Surface preparation: Peel ply.

The postbond humidity exposures data (82°C/wet) shows that in the case of precured laminates exposed to the dry or ambient conditions prior to bonding there is no degradation in performance. However, when the precured laminates were exposed to the hot/wet conditions prior to bonding, there was some degradation in properties after the post-bond humidity exposures. This shift in properties is attributed to the lowering of the stiffness of the composite and does not indicate a problem related to the adhesive system.

The failure modes in most cases was thin cohesive within the adhesive layer.

Similar trends are seen in the case of Hercules 3501-6/As 4 laminates. These data are shown in Table 6.

The precured composite substrate conditioning was also studied by the G1c (DCB double cantilever beam) test (Fig. 3). In this test the adhesive was cured between two precured skins and then tested in mode I. Test results on Ciba 6376 and the Hercules 3501-6 composite structures are shown in Tables 7a and 7b, respectively. These test results indicate that there is no degradation in the durability of the bonded structure even when the precured laminates are saturated with humidity prior to bonding. The failure modes for most of the G1c test specimens were found to be within the laminate. Table 8 shows the secondary bonding of composite sandwich structures.

The exposures to aircraft fluids were studied by exposing the FM® 300-2 bonded lap shear specimens to various fluids for 30 days at 23–60°C. The surface preparation for composite adherend was peel ply and the cured laminates were dried at 82°C for 1 h prior to bonding. Test results shown in Table 9 reveal that FM® 300-2 adhesive film bonded composite structures are not significantly affected by the exposures to Jet A, Skydrol or the Deicing fluid.

Secondary bonding data on CYCOM® 919, CYCOM 1827 and CYCOM 985 is shown in Table 10. The surface preparation used for these systems was peel ply and the humidity exposures were carried out by exposing individual lap shear specimens to 30 days at 71°C/100% RH. In the secondary bonding of 177°C cured graphite/epoxy or glass/epoxy laminates, FM 300-2 adhesive film shows excellent retention of its strength up to 93°C even after 30 days at 71°C and 100% relative humidity. Data

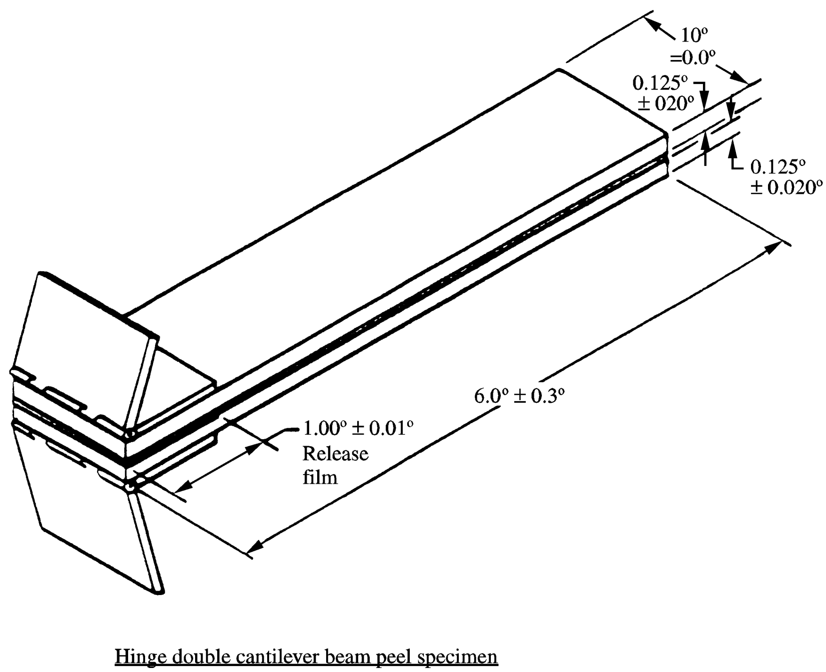

Fig. 3.

Table 7a
Secondary bonding of Ciba 6376 precured substrates with FM® 300-2 adhesive film. Effect of precured composite substrate conditioning on the double cantilever beam test

Property	Substrate conditioning	Test results		
		− 55°C	23°C	82°C/Wet
Hinged double cantilever test	30 days at @ 23°C	(20.8) 41.6	(15.5) 34.0	(15.9) 35.1
Crack energy (kg) Lbs	1 day @ 60°C/95% RH	(19.8) 42.2	(19.8) 43.5	(33.5) 73.6
	5 days @ 60°C/95% RH	(21.7) 47.9	(18.6) 41.0	(28.0) 61.7
Peel ply surface preparation	Dry 1 h @ 82°C	(20.5) 45.0	(15.1) 35.0	(20.0) 44.0
	Dry 1 h @ 121°C	(21.7) 47.7	(16.8) 37.0	(20.0) 44.0
	10 days @ 60°C/100% RH	(21.4) 47.1	(20.0) 44.0	(38.3) 84.3

Table 7b
Secondary bonding of Hercules 3501-6/AS4 composite with FM® 300-2 adhesive film. Effect of precured composite substrate conditioning on the double cantilever beam test

Property	Prebond substrate conditioning	Test results		
		− 55°C	23°C	82°C/Wet
Hinged double cantilever test	Dry 1 h @ 82°C	54.0 (24.5)	49.0 (22.3)	74.0 (33.6)
Crack energy (kg) Lbs	30 days @ 23°C	58.7 (26.7)	68.0 (30.9)	84.0 (38.2)
Peel ply surface preparation	10 days @ 60°C/100% RH	—	57.8 (26.3)	75.0 (34.1)

Table 8
Secondary bonding of composite sandwich structures. Effect of composite skin substrate conditioning on the flatwise tensile strength. FM® 300-2 adhesive film/Ciba 6376/3K70P Prepreg

Property	Substrate conditioning (face sheets)	Test results		
		− 55°C	23°C	82°C/Wet[a]
Flatwise tensile strength (MPa (psi))	Dry 1 h @ 82°C	4.7 (680)	5.1 (740)	2.2 (320)
Peel ply surface prepatation	10 days @ 60°C 95% RH	2.2 (314)	—	2.2 (315)

Note: Core: 0.128 g/cc density Nomex, 3.175 mm cell size. Failure mode: Core failure in all cases.

Table 9
Secondary bonding of Ciba 6376 Prepreg FM® 300-2M. Effect of fluid exposures on the carbon-carbon lap shear strength

Surface preparation	Substrate conditioning	Exposure conditions	Lap shear strength (mPa (Psi)@ 23°C)
Peel ply	Dry 1 h @ 82°C	30 days in Skydrol @ 60°C	18.8 (2725)
Peel ply	Dry 1 h @ 82°C	30 days in Jet A Fuel @ 23°C	21.4 (3100)
Peel ply	Dry 1 h @ 82°C	30 days in deicing fluid 23°C	20.8 (3020)
Peel ply	Dry 1h @ 82°C	No exposure (control)	20.7 (3000)

Note: Adhesive weight: 300 gm/m^2.

presented for 121°C cure laminates demonstrates good strength retention at room temperature. The service temperature for CYCOM® 919 is 71°C and, although bonded specimens were not tested at 71°C after humidity exposures, the strength retention is expected to be satisfactory.

4.2. *Effect of surface preparations*

The effect of surface preparation was studied by bonding the precured composite substrates with the FM® 300-2 adhesive film and then testing the bonded specimens in a lap shear test. The composite surface

Table 10
Bonding of precured thermosetting composite substrates with FM 300-2M (245 gm/m^2) adhesive

Precured composite substrate	Bonded specimen exposure	Lap shear strength (mPa (psi))		
		24°C	82°C	93°C
CYCOM 985/3K70P	None	23.7 (3440)	31.7 (4600)	30.6 (4430)
(Graphite/Epoxy)177°C cure	30 days at 71°C/100% RH	23.8 (3450)	22.8 (3300)	20.7 (3000)
CYCOM 1827/6781	None	16.0 (2320)	16.6 (2400)	16.2 (2350)
(glass/epoxy) 177°C cure	30 days at 71°C/100% RH	14.8 (2150)	13.8 (2000)	13.1 (1900)
CYCOM 919/3K70P	None	24.3 (3525)	— —	— —
(graphite/epoxy) 121°C cure	30 days at 71°C/100% RH	30.0 (4345)	— —	— —

Note: Adhesive weight: 245 gm/m^2.

Table 11a
Secondary bonding of ciba 6376 Prepreg with FM® 300-2M. Effect of substrate surface preparation on carbon-carbon lap shear strength

Composite surface preparation	Composite surface conditioning	Test results (mPa (psi))			
		− 55°C	23°C	82°C	82°C/Wet[a]
Peel ply	Dry 1 h @ 82°C	18.5 (2680)	20.7 (3000)	22.9 (3315)	27.9 (4050)
Peel ply/hand sand	Dry 1 h @ 82°C	17.4 (2525)	20.1 (2910)	29.9 (4330)	29.1 (4215)
Peel ply/grit blast	Dry 1 h @ 82°C	25.7 (3725)	30.6 (4435)	31.5 (4565)	31.5 (4560)

[a]Wet conditioning: 30 days at 71°C/100% RH.

Table 11b
Secondary bonding of Ciba 6376 precured substrates with FM® 300-2 adhesive film. Effect of composite surface preparation on double cantilever beam testing

Test	Composite surface preparation	Test results		
		− 55°C	23°C	82°C/Wet
Hinged double cantilever test	Peel ply	45.0 (20.45)	35.0 (15.91)	44.0 (20.0)
Crack energy (kg) Lbs	Peel ply/hand sand	45.5 (20.68)	39.0 (17.73)	47.9 (21.77)
Substrate dried 1 h @ 180	Peel ply/grit blast	73.6 (33.45)	49.0 (22.27)	80.4 (36.54)

Note: Wet conditioning: 30 days @ 71°C/100% RH.

preparations studied included (a) dry Nylon peel ply, (b) Peel ply/hand sand, and (c) Peel ply /grit blast for the Ciba 6376/3K 70P prepreg system. The surface preparation for the Hercules 3501-6 carbon/epoxy prepreg included (a) peel ply and (b) peel ply/hand sand.

The test results on the Ciba 6376 prepreg are shown in Table 11a. These data show that peel ply followed by the sand blasting offers the best overall mechanical performance and the dry peel ply is the worst. The effect of the surface on the hinged double cantilever beam test for the Ciba 6376 prepreg is shown in Table 11b. These test results also show that grit blast in addition to peel ply provides the most durable bond. In the case of the Hercules 3501-6, there was no significant difference between the peel ply versus the peel ply/hand sand surface preparation. Either of these two surface preparations provided good performance under both the dry and the hot/wet conditions. The test data are shown in Table 12.

The failure modes for most of the lap shear tests ranged from cohesive in the case of grit blast prepared substrates to mostly interfacial between the adhesive and the laminate for the peel plied surface preparation.

In the case of Hercules 8551-7 epoxy/glass and 8551-7 epoxy/graphite laminates the surface preparations included (a) solvent wipe only, (b) hand sanding/solvent wiping, and (c) sandblast/solvent wipe. No peel ply was used.

Laminate layup was $(\pm 45, \pm 90, 0)_2$ for both the glass and the graphite substrates.

Humidity exposures were carried out by exposing individual lap shear specimens to 30 days at 71°C/100% RH.

Table 12
Secondary bonding of Hercules 3501-6/3K70P Prepreg with FM® 300-2 adhesive film. Effect of composite surface preparation on carbon-carbon lap shear strength

Surface preparation	Lap shear strength (mPa (psi))			
	− 55°C	23°C	82°C	82°C/Wet
Peel ply	22.6 (3280)	28.7 (4160)	30.4 (4400)	30.2 (4380)
Peel ply/hand sand	22.5 (3270)	28.6 (4145)	29.1 (4220)	33.0 (4785)

Note: Substrate dried at 82°C for 1 h prior to bonding.

Table 13
Effect of surface preparation on the LSS of FM 300-2K, 390 gm/m^2 adhesive bonded precured Hercules 8551-7 laminates
Lap shear strength (mPa (psi)

Test temp. (°C)	Hercules 8551-7/ graphite	Hercules 8551-7/ glass	Surface preparation
− 55°C	13 (1885) (AL)	10.8 (1565) (AL)	
24°C	19.6 (2845) (AL)	16.3 (2365) (AL)	Solvent wipe only
82°C	21.1 (3065) (AL)	19.0 (2760) (AL)	
82°C/Wet	15.1 (2190) (AL)	11.6 (1685) (90% A, 10% C)	
− 55°C	18.2 (2645) (AL)	14.8 (2145) (LF)	
24°C	23.7 (3435) (AL)	17.9 (2590) (LF)	Hand sand (320 grit sandpaper)/ solvent wipe
82°C	25.6 (3715 (AL)	18.6 (2620) (AL)	
82°C/Wet	17.8 (2575) (50%A, 50%C)	12.8 (1855) (90%A, 10%C)	
− 55°C	22.6 (3285) (LF)	12.7 (1835) (LF)	
24°C	27.5 (4005) (LF)	14.8 (2150) (LF)	Sandblast/solvent wipe
82°C	27.5 (3985) (LF)	14.7 (2130) (LF)	
82°C/Wet	19.9 (2885) (50%A, 50%C)	11.0 (1600) (40%A, 60%C)	

Note: Laminate: Hercules 8551-7 Uni-laminate ($\pm$ 45, $\pm$ 90, 0)$_2$.
Cure cycle: 1.7°C/min to 120°C, 60 min at 120°C, /28 mPa.
Failures modes: AL = Adhesive to laminate, C = Cohesive, LF = Laminate failure, A = Adhesive.

Test results are shown in Table 13. The data on the secondary bonding of Hercules 8551-7/graphite laminates show that sandblast followed by solvent wiping provided the best overall strength in the bonded structure. Lap shear values from the hand sanded specimens were on an average 10–20% lower than the sandblasted specimens. The solvent wipe only was found to be the least effective method for surface preparation of the Hercules 8551-7/graphite laminates. The failure modes for both solvent wipe as well as hand sanded surface preparations were predominantly adhesive to laminate, whereas for the sandblast prepared specimens, the failure mode was mostly laminate failure. The laminate failure was mostly in the top plies of the $\pm$ 45° layup.

However, in the case of Hercules 8551-7/glass precured laminates, it was more difficult to discern the effect of surface preparation on the lap shear strength. Due to the laminate lay-up, laminate failure was the predominant failure mode for both sanded and sandblasted specimens. Both solvent wipe only and hand sanded specimens provided higher overall shear values as coMpared to the sandblasted specimens.

Results from this study show that peel ply followed by grit blast provides the best overall durability of the

Table 14
Cocure and secondary bonding of ciba M20/3K70P prepreg with FM® 300-2M adhesive film. Cured 2 h at 121°C under vacuum only cure conditions

Test	Test condition	Test results (mPa (psi))
Precured substrates carbon–carbon lap shear strength (Grit blast/MEK wipe surface preparation)	23°C	27.0 (3915)
	71°C	27.4 (3975)
	30 days at 71°C/100% RH then test @	
	23°C	26.9 (3900)
	71°C	26.9 (3905)
Cocured flatwise tensile testing	23°C	3.7 (530) (core failure)
BMS 8-124. Class 4, Type 5 Core	71°C	3.6 (525) (core failure)
	93°C	4.5 (645) (core failure)
Cocured beam shear BMS 8-124, Class 4, Type 5 Core	23°C	3.6 (515)
	71°C	3.3 (475)
	93°C	2.9 (415)

bonded structure. Where it is not practical to grit blast, light hand sanding should be considered in addition to the peel ply.

4.3. Repair applications/vacuum curing

To study the utility of the FM 300-2 adhesive film for repair type bonding applications, a series of studies were undertaken to evaluate the performance of this adhesive under vacuum only cure conditions. These studies were conducted with the 121°C curing Ciba M 20 epoxy/carbon prepreg and the FM® 300-2 adhesive film in both co-cure and the secondary bonding conditions. Some work was also conducted with the 177°C curing Ciba 637 3K70p prepreg. The cure cycle for the vacuum only cure included a 2°C/min heat up to 121°C and a hold temperature of 90 min at 121°C.

In the secondary bonding studies, the surfaces were prepared by grit blasting followed by solvent wiping with methyl ethyl ketone for the Ciba M20 prepreg and the peel ply for the Ciba 6376 material. The lap shear panels were then bonded under vacuum at 121°C and tested before and after humidity exposures. The test results are shown in Tables 14 and 15. The data on the secondary bonding of both these prepregs show that the FM® 300-2 adhesive film provides excellent bondability under vacuum only cures and shows no drop-off in mechanical strength even after 30 days exposure to 71°C at 100% RH.

The cocure studies were conducted by building sandwich panels comprising of 0.128 g/cc Nomex core, FM® 300-2 adhesive film and eight plies of Ciba M 20 3K70P prepreg on each side of the core. The adhesive and the prepreg were co-cured under vacuum at 121°C and then tested as flatwise tensile and beam shear specimens. Test results (Table 14) show that from 23°C to 93°C test temperatures the flatwise tensile strength remains unchanged with 100% core failures. Similar trend was seen with the beam shears with results ranging from 3.55 mPa (515 psi) at 23°C to 2.86 mPa (415 psi) at 93°C with core buckling as the predominant mode of failure.

These vacuum only cure test results indicate that FM® 300-2 adhesive film can be used as a repair adhesive and provides good performance in both co-cure and the secondary bonding applications.

4.4. Thermoplastic substrates

For the secondary bonding of thermoplastic substrates (PEEK and KIII polymer), the knit supported adhesive film was used. This data (Table 16a) shows that the 121°C curing adhesive provides the same performance as 177°C cured FM® 300 adhesive over the temperature range of 55–150°C in bonding of PEEK laminates. The composite bonding data on DuPont's KIII thermoplastic composite substrate adhesive is shown in Table 16b. Strength retention is excellent up to 121°C dry and 82°C after humidity exposures.

Table 15
Secondary bonding of Ciba 6376 Prepreg with FM® 300-2 adhesive film carbon-epoxy prepreg. Effect of vacuum only cure

Test	Composite surface preparation	Composite substrate condition	Test temperature	Test results (mPa (psi))
Carbon–carbon lap shear	Peel ply	Dry 1 h @ 82°C	− 55°C	18.4 (2670)
Carbon–carbon lap shear	Peel ply	Dry 1 h @ 82°C	− 23°C	20.0 (2895)
Carbon–carbon lap shear	Peel ply	Dry 1 h @ 82°C	82°C	20.6 (2980)
Carbon–carbon lap shear	Peel ply	Dry 1 h @ 82°C	82°C/Wet[a]	22.8 (3300)

[a]Bonded specimens exposed for 30 days at 71/100% RH.

Table 16a
Comparative data between FM 300-2 and FM 300 adhesives secondary bonding of thermoplastic composite substrate

Adhesive system	Precured composite Substrate	Lap shear strength (mPa (psi))			
		− 55°C	24°C	149°C	82°C Wet[a]
FM 300-2K (392 gm/m^2)	PEEK (APC-2)	21.2 (3075)	25.2 (3655)	13.4 (1940)	22.2 (3220)
FM 300K (392 gm/m^2)	PEEK (APC-2)	21.4 (3100)	25.2 (3650)	12.6 (1820)	21.8 (3165)

Note: Surface preparation: Plasma etched.
Cure temperature: FM 300-2: 90 min at 121°C, 0.28 mPa, heat up 1.7°C/min.
FM 300: 60 min at 177°C, 0.28 mPa, heat up 1.7°C/min.
[a]Individual coupons exposed 30 days at 60°C and 100% RH.

Table 16b
Bonding of DuPont KIII laminate substrate with FM 300-2K (392 gm/m^2) adhesive. Effect of postbond humidity exposure

Bonded specimen Exposure	Lap shear strength (mPa (psi))		
	24°C	82°C	121°C
None	16.7 (2420) (70% C) (30% LF)	17.7 (2570) (60% C) (40% LF)	16.1 (2340) (85% A) (15% C)
30 days at 71°C 100% RH	17.9 (2600) (LF)	17.9 (2600) (50% C) (50% LF)	10.7 (1550) (100% C)

Note: Surface preparation: Sandblast/solvent wipe
LF = Laminate Failure; C = Cohesive; A = Adhesive failure.

Table 17
Composite bonding data FM 300-2 adhesive co-cured with CYCOM 985/AS-4 Prepreg system

Adhesive	Bonded specimen exposure	Wide area LSS (mPa (psi))	
		24°C	104°C
	None	17.3 (2510) (LF)	15.0 (2180) (LF)
FM 300-2M (147 gm/m^2)	30 days at 60°C/ 100% RH	22.4 (3250) (LF)	21.7 (3140) (LF)

Note: Cured laminate thickness: 3.56 mm. LFD: Laminate failure.

4.5. Co-cure studies

For co-cure and surfacing studies, adhesive film with a mat carrier was co-cured with 177°C curing prepreg. After curing, the laminate was cut into one inch wide double notched wide area lap shear coupons. The test results are tabulated in Table 17.

Results were excellent under both dry and wet conditions over the temperature range of 24°C to 104°C. The mode of failure in all cases was cohesive within the laminate plies. The surface ply studies were done with mat supported (147 or 245 gm/m^2) adhesive films. Although 147 gm/m^2 adhesive film provided good surfacing properties, even better surface smoothness was obtained with the heavier 245 gm/m^2 adhesive film. Use of surface ply not only provides smooth ready to paint surfaces but also can reduce moisture penetration into composite skins.

4.6. Interleafing applications

For interleafing applications in composites, we have developed a low flow adhesive. Designated as FM® 300-2 Interleaf, it is a 121°C curing, low flow, modified epoxy based adhesive. Because of its low flow, and lower cure temperature, it resists intermixing with the prepreg resin and provides a high strain, low modulus layer between the plies of the laminate. This results in better stress distribution within the laminate and higher impact strength [4].

The interleaf adhesive can be co-cured with most epoxy prepreg systems and provides excellent composite surfacing properties. Because of its lower flow, the interleaf adhesive provides even better surfacing properties than the standard adhesive.

The effect of humidity exposure on the lap shear strength of mat supported interleaf adhesive is shown in Table 18. Data on 177°C cure mat supported FM® 300 Interleaf is also included for coMparative purposes. These results show that the interleaf adhesive can be co-cured even with a high flow 177°C curing epoxy prepreg system CYCOM® 985-1 and provides performance similar to FM® 300 Interleaf adhesive.

5. Summary

Performance of FM® 300-2, a state-of-the-art 121°C curing epoxy based adhesive for various metal and composite bonding applications has been described. This adhesive can be cured at 121°C to 177°C and provides similar physical and mechanical properties irrespective of the cure temperature. Due to its low moisture absorption and high glass transition temperature, this adhesive

Table 18
Co-cure data - FM 300-2 interleaf or FM 300 interleaf adhesives co-cured with 177°C curing epoxy prepreg systems

Adhesive	Prepreg system	Bonded specimen exposure	Wide area lap shear strength (mPa (psi))	
			24°C	104°C
FM 300-2M Interleaf (245 gm/m^2)	CYCOM 985/AS-4 (graphite/epoxy controlled flow system)	None	20.8 (3010)	21.7 (3140)
		30 days at 71°C/100% RH	17.2 (2515)	18.8 (2730)
FM 300M Interleaf (245 gm/m^2)	CYCOM 985/AS-4 (graphite/epoxy controlled flow system)	None	18.8 (2720)	17.9 (2600)
		30 days at 60°C/100% RH	14.1 (2040)	19.0 (2760)
	CYCOM 985-1/AS-4 (graphite/epoxy high flow system)		21.4 (3100)	19.8 (2875)
		30 days at 60°C/100% RH	23.4 (3390) (LF) (LF)	19.3 (2800)

Note: Cured laminate thickness = 3.56 mm. Failure mode: Laminate failure at 24°C and cohesive at 104°C.

shows excellent performance when bonding to wet Nomex honeycomb and in bonding precured thermoset and thermoplastic composite substrates. It can be co-cured with most 121°C or 177°C curing epoxy prepreg systems and provides excellent bonding and surfacing properties. In addition, performance of a low flow grade of FM® 300-2 Interleaf adhesive has been described for applications such as surfacing, interleafing and repair of composite structures.

References

[1] Politi RE. SAMPE International Technical Conference, 1987; 19:36.
[2] Kohli DK. Proceedings of 10th International European Chapter Conference of the SAMPE 1989;55:239.
[3] Kohli DK. 39th International SAMPE Symposium, 11–14 April 1994.
[4] Hirschbuehler KR. SAMPE Quarterly, 1985;17.