## Materials for Heated Head Automated Thermoplastic Tape Placement <sup>†</sup>

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### Abstract

NASA Langley Research Center (LaRC) is currently pursuing multiple paths to develop out of autoclave (OOA) polymeric composite materials and processes. Polymeric composite materials development includes the synthesis of new and/or modified thermosetting and thermoplastic matrix resins designed for specific OOA processes. OOA processes currently under investigation include vacuum bag only (VBO) prepreg/composite fabrication, resin transfer molding (RTM), vacuum assisted resin transfer molding (VARTM) and heated head automated thermoplastic tape placement (HHATP). This paper will discuss the NASA Langley HHATP facility and capabilities and recent work on characterizing thermoplastic tape quality and requirements for quality part production. Samples of three distinct versions of APC-2 (AS4/PEEK) thermoplastic dry tape were obtained from two materials vendors, TENCATE, Inc. and CYTEC Engineered Materials\*\* (standard grade and an experimental batch). Random specimens were taken from each of these samples and subjected to photo-microscopy and surface profilometry. The CYTEC standard grade of APC-2 tape had the most voids and splits and the highest surface roughness and/or waviness. Since the APC-2 tape is composed of a thermoplastic matrix, it offers the flexibility of reprocessing to improve quality, and thereby improve final quality of HHATP laminates. Discussions will also include potential research areas and future work that is required to advance the state of the art in the HHATP process for composite fabrication.

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## **1.0 Introduction**

The NASA Langley Research Center's heated head automated thermoplastic tape placement (HHATP) facility utilizes a high-rail gantry system providing 1.8 meter (~6 feet) of linear motion, a 1m x 1m (~3ft x 3ft) flat table controlled by X and Y direction stepper motors, and a heated tape placement head built by Accudyne, Inc. (Newark, DE) under a Small Business Innovative Research (SBIR) Phase II contract. The HHATP process is fully automated with process and machine control software developed by Accudyne engineers. The coordinated gantry/head motion together with the on-head polymer process enables the fabrication of opensection flat laminates without requiring an autoclave. The deposition head preheats the thermoplastic tape and the pre-placed laminate to the melting temperature, and then welds them together under heat and pressure until the composite material is healed and consolidated in place [1]. Figure 1(a) shows the HHATP facility and (b) a close-up front view of the placement head.



(a)

(b)

Figure 1. Pictures of the Langley HHATP facility (a) and close-up front view of Accudyne, Inc. heated head (b).

Figure 2(a) shows the processing elements used in composite fabrication including: 1) four MAPP gas combustion torches to preheat the already-placed composite layers to near the process temperature, 2) a mini gas torch to rapidly heat both tape surfaces at the contact point to the process temperature (the heating is controlled to within 10°C by two cascaded multiple input/single output controllers), 3) the 7.62cm (3in) tape feeder with nip, clamp, and cut mechanisms, 4) heated conformable compactors capable of applying force through a compaction range of 12mm at 450°C. A line compactor can apply 800N (~180 lb) and an area compactor can apply 1330N (and 300lb) over an area of 11.4cm x 10cm (4.5in x 4in), and 5) chilled area conformable compactors that can apply forces of 2200N and 1330N (~500 and 300lb), respectively, at 10°C over a range of motion of 12mm and an area of 12.7cm x 10cm (5in x 4in). Figure 2(b) shows the detail of the conformable compactors capable of producing the approximate contour of a basketball, i.e. double curvature with a radius of about 12 cm (4.8 in).

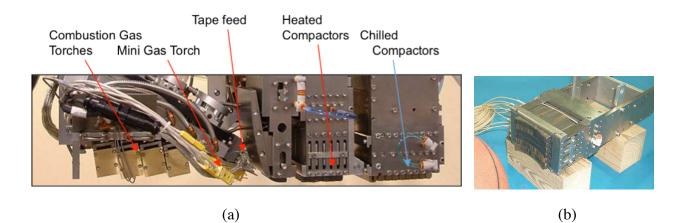


Figure 2. Close-up pictures of process elements including torches, tape feeder mechanism, and heated and chilled compactors (a) and heated area compactor showing conformance similar to basketball contour (b).

Recent work shows that the HHATP process alone delivers ~85 to 90% of the typical composite mechanical properties (open hole compression, 0° short beam shear, 0° Flex) delivered when the HHATP process is followed by a standard autoclave process cycle [2]. Because the HHATP process is relatively rapid, there is insufficient time to remove voids, generate the full intimate contact required for interply healing, or to wet out dry fibers. Poor quality tape, that contains voids or surface roughness that limits intimate contact between plies, will produce substandard composites required for wing and fuselage built up structure, or for composite tanks. Thus, the highest priority research area to advance the technology readiness level of this process is improving tape quality [2].

Researchers in NASA Langley's Advanced Materials and Processing Branch, using the tape line shown in Figure 3, have made experimental 3-in-wide dry tape [4, 5] from several thermoplastic polyimides. Further optimization of the tape fabrication process should produce tapes of the quality needed for the HHATP process. LaRC researchers have considerable experience developing high performance/high temperature resistant thermoplastic polyimides [3]. These are typically totally amorphous with high glass transition temperatures (Tg) and show very good potential as matrix resins for carbon fiber composites.



Figure 3. Langley dry tape processing line for thermoplastic polyimides.

# 2.0 Experimentation

## 2.1 Materials

Samples of three distinct versions of AS4/PEEK thermoplastic dry tape were obtained from two materials vendors, TENCATE, Inc. (Union City, GA) and CYTEC Engineered Materials (Havre de Grace, MD). The TENCATE AS4/PEEK tape was received as a 30.5 cm (12in) wide roll approximately 37 m (120 ft) in length. CYTEC supplied APC-2 standard tape as several 7.62 cm (3.0 in) wide rolls approximately 122 m (400ft) in length as well as ~3ft of "Experimental" tape. A 6.0 mg specimen was obtained from each sample of preimpregnated carbon fiber tape and analyzed in differential scanning calorimetry (DSC) at a heating rate of 10°C/min to determine the degree of crystallinity and thermal history of the as-received rolls of dry tape. All three tape samples were also potted, polished, and photo-micrographed in order to examine their cross sections.

### **2.2 Profilometry**

Two 7.62cm x 7.62cm (3in x 3in) samples from eachroll of tape were sent to profilometer vendors for independent analysis of the material surface roughness. Each of the five vendors (Solarius Development, Sunnyvale, CA; FRT, Somers, CT; Nanovea, Irvine, CA; Zygo, Middlefield, CT; ZETA, San Jose, CA) employed one of their non-contact profilometers to report the arithmetic mean surface roughness, Sa, and the maximum peak-to-valley height, Sz, measured from at least two 25mm x 25mm (1in x 1in) areas of the dry tape supplied to them for analysis.

## **3.0 Results and Discussion**

### **3.1 Differential Scanning Calorimetry (DSC)**

The results of the dynamic DSC scans for each of the three tape samples are shown in Figure 4.

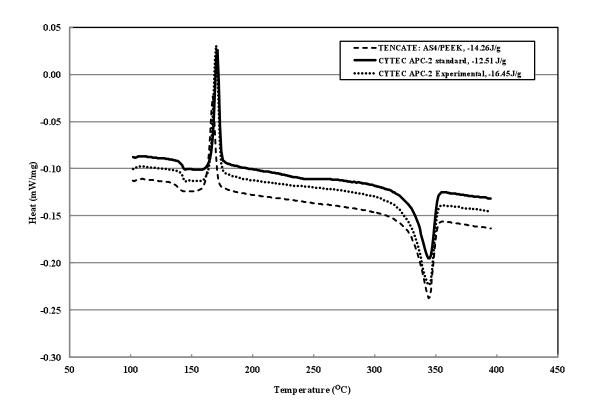


Figure 4. Dynamic DSC of pristine dry tape samples and resulting heat of crystalline melt.

Comparison of the energy of crystalline melt of the TENCATE AS4/PEEK (-14.26 J/g) and the APC-2 "Experimental" grade (-16.45 J/g) shows both have higher crystalline content than the CYTEC standard APC-2 tape (-12.51 J/g). The more crystalline material would typically have been subjected to a longer annealing time above the thermoplastic Tg but below the crystalline  $T_m$  during processing at the supplier. A 6.0 mg specimen of the CYTEC APC-2 standard tape that was heated to 280°C and then immediately quenched gave a measured crystalline melt of -12.55 J/g.

## **3.1 Profilometry Results**

The 3-D images of the surfaces of the tape samples are displayed in Figures 5-7, and the numerical profilometry results are reported in Table 1. In spite of the variations in Sa reported by the separate vendors, the results indicate that both the TENCATE AS4/PEEK and the CYTEC "Experimental" grade tape are significantly smoother than the CYTEC "Standard" APC-2 material. This is important because improved surface contact between smoother plies results in both higher lay down rates (improved process through-put) as well as better quality CFRP laminates. A process model by Lamontia, et.al. [6] includes a parameter for the average surface

roughness and shows these effects. A FRT Microprof® 100 Profilometer (Somers, CT) that has been purchased and delivered to LaRC will be used to extend the measurements presented in Table 1.

			Number of sample locations taken from	average surface roughness, Sa	average of Maximum height, Sz	average skewness,
Sample#	Description	Company	eachspecimen	( <sup>µ</sup> m)	(µ <b>m)</b>	Ssk
A1	CYTEC Standard	SOLARIUS	3	4.32	37.53	N/R
A2	CYTEC Standard	FRT	З	9.21	57.9 <i>7</i> 5	-0.114
B1	CYTEC Experiment:	I NANOVEA	З	0.654	15.714	0.112
B2	CYTEC Experiment:	al ZYGO Corp	2	0.204	11.017	N/R
Q	<b>TENCATE</b> Standar	A TET A	2	2.291	5.78	0.0253

Table 1: Preliminary Surface Profilometry Results.

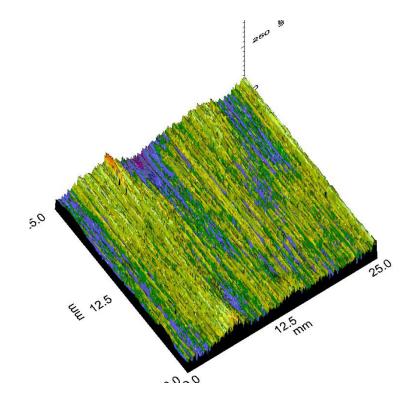


Figure 5. A 3-D surface profile of CYTEC APC-2 "Standard" tape (taken by FRT of AMERICA).

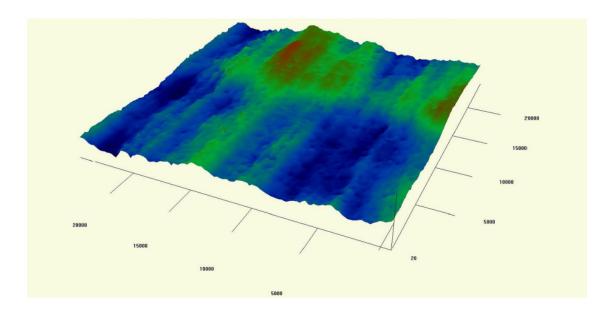


Figure 6. A 3-D surface profile of TENCATE AS4/PEEK tape (taken by ZETA Instruments).

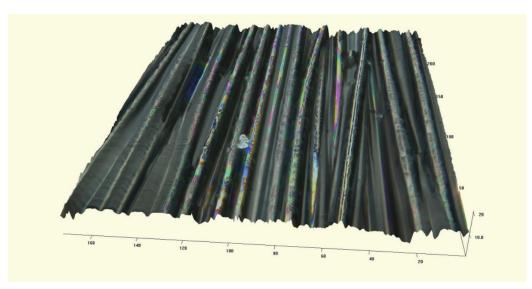


Figure 7. A High resolution 3-D surface image of TENCATE AS4/PEEK tape (taken by ZETA Instruments).

## **3.2 Through Thickness Quality Evaluation**

In addition to the surface analysis Figures 8 and 9 indicate that the TENCATE tape and the CYTEC "Experimental" tape, respectively, are void free and contain a uniform dispersion of AS-4 fibers and thermoplastic resin. These photomicrographs are of specimens taken from the

centers of the as-received rolls are consistent with the specimens taken from all regions of the tapes provided by both suppliers. Figure 10 displays a cross-sectional micrograph of a specimen taken from the center of the 7.62cm wide CYTEC "Standard" APC-2 tape. This material consistently contains random voids, splits and resin-rich regions across the width. The HHATP process cannot overcome these deficiencies in quality. The TENCATE tape and the "Experimental" grade CYTEC tape appear to be more suitable to the HHATP process.

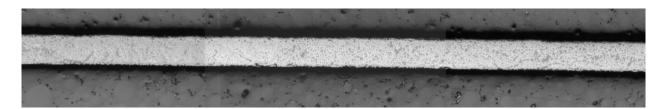


Figure 8. Photo-microscopy of cross-section of CYTEC APC-2 "Experimental" tape, 50X.

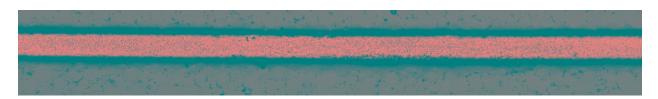


Figure 9. Photo-microscopy of cross-section of TENCATE AS4/PEEK tape, 50X.

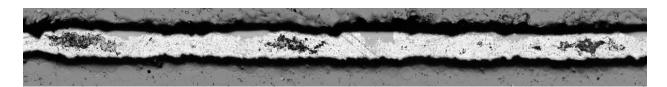


Figure 10. Photo-microscopy of cross-section of CYTEC APC-2 "Standard" tape, 50X.

### **3.3 Thermoplastic Tape Processing Parameters**

The thermoplastic polyimides developed at NASA Langley show very good potential as matrix resins. For example, carbon fiber (IM7) composites prepared with LaRC 8515 provide very good properties [7, 8]. LaRC 8515 is an amorphous polyimide with at Tg of ~270°C. While typically considered a problem when processing, a high Tg with no crystalline melt temperature (Tm) can actually be beneficial when considering the HHATP process. For example, the PEEK matrix material used in the APC2 tape has a Tg ~ 140°C and a Tm ~ 340°C. Therefore, the previously placed composite and the incoming tape surfaces must be heated above the Tm and then held under the chilled compaction pressure until the composite is cooled below Tg. For

amorphous polymers with no Tm, the tapes need only to be heated above Tg to a temperature needed to produce the required flow. This temperature is relatively high, but it is easily achieved in the HHATP process because torches and heated compactor have ample heating capability even at relatively high lay down rates. However, with low Tg resins like PEEK, the cooling requirements can easily exceed the current equipment capability when faster lay down rates are attempted. A high Tg matrix requires less cooling to freeze the matrix in place, so faster lay down rates are possible without modification to the Langley HHATP facility. A potential area for future work is the development of high quality tape from high Tg polyimide thermoplastics, followed by fabrication of these tapes into composites using HHATP.

#### 4.0 Future Work

The existing LaRC tape line will be converted to heal as-received standard grade tape CYTEC APC-2 tape. Temperatures, pressures and times needed to heal the voids and splits as well as to lower the surface roughness will be determined using a heated press. An NDE method such as through transmission c-scan or Photometry will be employed and validated as a tool to quickly determine the improvements made in the quality of the tapes. Optimized APC-2 tape as well as the TENCATE tape will be placed using the LaRC HHATP facility and resulting composites will be tested. The in-house thermoplastic tape fabrication facility will be used to prepare dry tape from LaRC 8515 and additional experimental resins developed at NASA so resulting composites can be fabricated and evaluated.

#### 5.0 Conclusions

#### 5.1 Summary

NASA LaRC has been investigating the use HHATP for aerospace grade composite fabrication and has discerned that the biggest hurdle to overcome in HHATP is poor quality tape. In this work, three versions of AS4/ PEEK tape were acquired and characterized to assess their potential use with the HHATP process. Cytec standard grade AS4/ PEEK tape was determined to have a large void content and poor surface quality that would limit the quality of any composite made by HHATP using this tape. The HHATP process cannot consolidate the voids during the short processing times associated with this type of process. However, the Cytec Experimental grade tape that was evaluated was determined to be essentially void free and had a significantly improved surface quality that would lend itself to successful HHATP. The AS4/ PEEK tape acquired from TENCATE also was void free and had a better surface quality than the standard grade Cytec tape. With the proper quality tape, HHATP can produce aerospace grade quality OOA in-situ composite parts.

## 6.0 References

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