LIMITATIONS ON MECHANICAL PROPERTIES IN THERMOPLASTIC LAMINATES FABRICATED BY TWO PROCESSES: AUTOMATED THERMOPLASTIC TAPE PLACEMENT AND FILAMENT WINDING

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SUMMARY

This paper contrasts challenges in developing two thermoplastic in situ consolidation processes: filament winding and fiber placement. The filament winding section details the mechanical properties of AS-4/PEEK and AS-4/PEKK $[90_m/0]n$ cylinders fabricated by combining filament winding for 90° plies with tape placement for 0° plies. Properties show the negative impact of void and ply waviness defects on compression and interlaminar shear strengths as well as on cylinder structural collapse under hydrostatic compression loading. Also, performance equivalent to autoclave baselines has been achieved with good laminate microstructure.

AS-4 and IM-7/PEEK, PEKK, and polyimide flat laminates have been fabricated via thermoplastic tow and tape placement (ATP) with low voids and waviness, but their OHC strengths still do not reach those from autoclaved laminates. Why? Full polymer matrix crystallinity appears never to have been achieved from in situ processing. Without full crystallinity, the full resin modulus is never attained, and compression properties suffer. A new hypothesis tying the loss in fiber-placed property performance with inadequate matrix crystallinity is advanced, and a new area of development for materials suitable for in situ fiber placement is proposed.

1. INTRODUCTION

Our development of thermoplastic filament winding and fiber placement has been focused by four programs. First, the 1988 DARPA Advanced Submarine Technology (AST) Program demonstrated successful thermoplastic cylinders fabricated with in situ filament winding for hoop plies and tape placement for axial plies. Second and third, the 1993 RAPTECH-PMC and 1995 RAPTECH-ACM Programs demonstrated thermoplastic tow and tape placement of AS-4/PEEK and AS-4/PEKK flat laminates with an in situ consolidation head on a fiber placement machine and then a gantry. Fourth, the 1994-99 NASA High Speed Research Program extended fiber placement development with polyimide material systems.

The in situ processing promise of cost reduction in large part fabrication has been calculated [1,2]. With favorable economics, the goal was to demonstrate process viability by proving that excellent properties can be achieved in a useful shape. This quest continues for in situ placed flat or moderately contoured laminates, with the current focus to meet 327 MPa (47.4 Ksi) Open Hole Compression (OHC) strength property targets set by autoclave-cured or compression-molded laminate tests.

2. EFFECTS OF DEFECTS IN CYLINDERS

Prior to the AST program, David Taylor Research Center (now NSWC-CD) contracted the fabrication of thermoset autoclaved cylinders [3]. Disappointing performance resulted when hoop waviness formed during autoclave curing of the thick laminates. Better strains at failure resulted from Hitco sequentially-cured cylinders having low waviness. Later, DuPont fabricated a number of $[0/90_m]n$ cylinders with a fully automated in situ process combining tape-placed axial plies and filament wound hoop plies. Since the in situ process by nature sequentially consolidates each ply layer individually before placing the next, low waviness and excellent properties naturally result from the thermoplastic in situ process.

Elevated void content can also be problematic. Figure 2.1 organizes the thinking around how these two defects that can act apart or synergistically. This section will show that void content can lower compression properties alone, or in concert with waviness acting though a low shear modulus and strength.

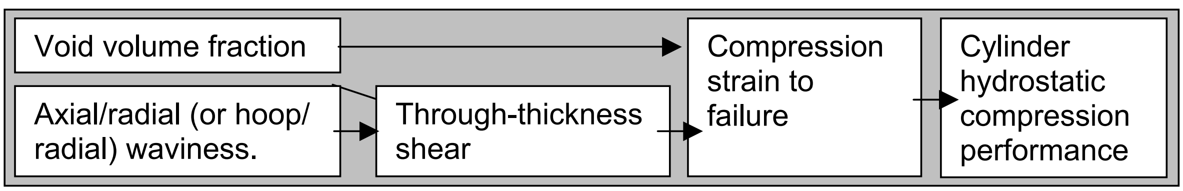

Figure 2.1 The effect of void content and waviness, through shear, on compression strain-to-failure and then to cylinder hydrostatic compression load carrying capability

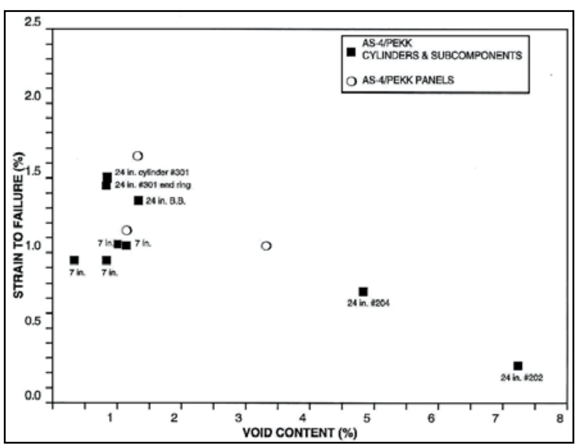

Figure 2.2 Compression Strain-to-Failure for AS-4/PEKK Cylinders and Laminates

The correlation between strain-to- failure and void content is evident from Figure 2.2, which shows that the compression strain-to-failure drops for low ply waviness carbon/PEEK and PEKK cylinders with increases in void content from 1% or less to 5% to 7%.

Excessive layer waviness also impacts compression load carrying capability. Wavy fiber layers transform simple compression to combined compression and shear. If the composite shear strength and modulus is insufficient to channel the force along the wavy fiber, a loss in compression strength results. Figure 2.3 details the negative effect of voids on shear strength. Figure 2.4 shows that elevated shear strengths are required to maintain fiber-dominated failure modes in thick composite cylinders.

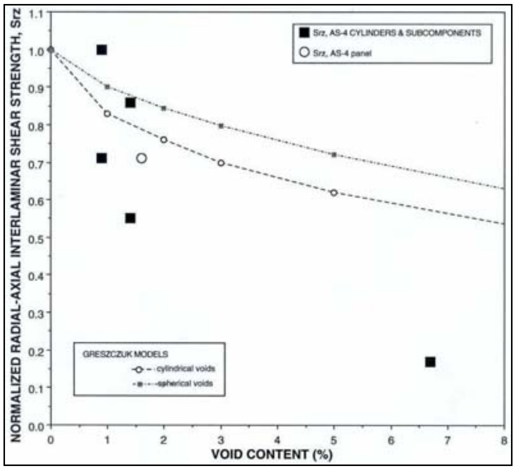

Figure 2.3 Void content negatively affects interlaminar shear strength

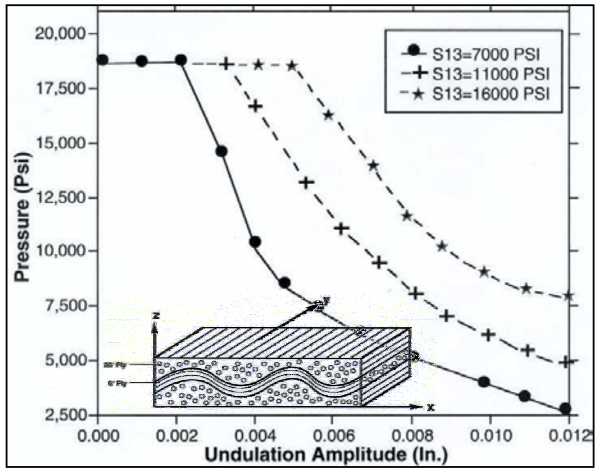

Figure 2.4 Influence of waviness undulation amplitude on collapse pressure of AS-4/carbon PEEK and PEKK cylinders

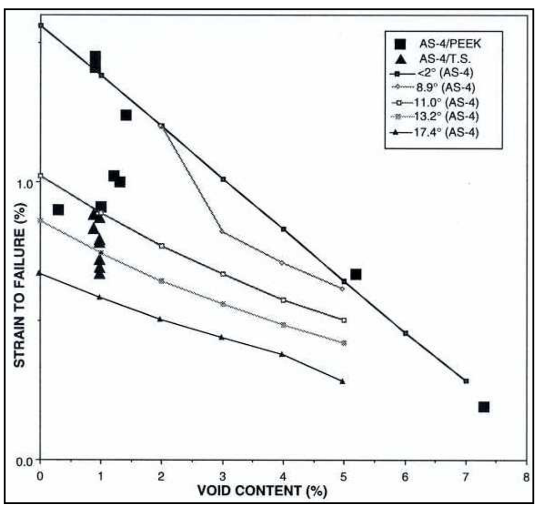

Figure 2.5 combines the synergistic effect of voids and waviness on strain-to-failure. Voids alone reduce compression strain-to-failure in cylinders with waviness less than 2°. Increasing void content acts to reduce shear strength, and through waviness, also lowers the compression strain-to- failure.

Figure 2.5 Void content directly lowers compression strain-to-failure, or in addition, lowers shear strength. Layer waviness, acting through the depressed shear strength, lowers the compression strain-to-failure even further than voids alone.

3. TOW AND TAPE PLACEMENT

In tow and tape placement, excessive fiber waviness and elevated void content has been overcome, as shown in Figure 3.1 for an IM-7/PIXA tape-placed laminate fabricated during the NASA HSR program. However, there has generally been only an 85% Open Hole Compression strength translation compared with autoclaved laminates of the same material system [5, 6], as shown in Figure 3.2. What is the cause?

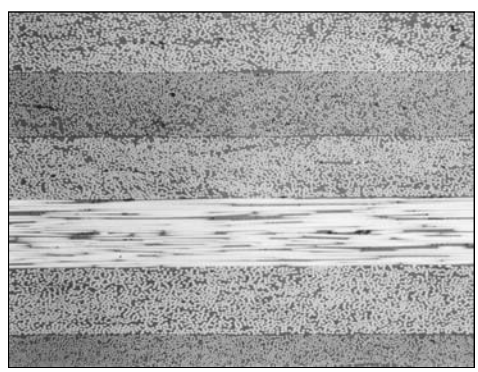

Figure 3.1 A quasi-isotropic laminate fabricated from IM-7/PIXA tape with an in situ consolidation tape placement process is void and waviness free

One possibility is that the level of crystallinity was inadequate in the placed laminates. That is, semi-crystalline polymers like PEEK, PEKK, and

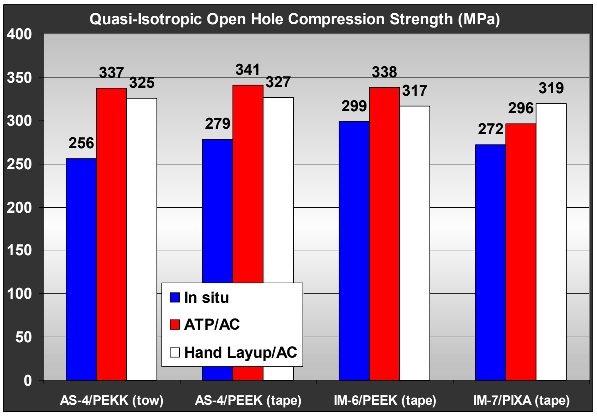

Figure 3.2 OHC for PEKK, PEEK & PIXA

PIXA only develop their full level of crystallinity given adequate time at temperature during cooling to crystallize out of the melt, for example like times characteristic of autoclave processing. This time is not duplicated with in situ placed laminates that cool at >100°C/second.

Two PEKK laminates were fabricated, one on a heated aluminum tool and another on an unheated insulated tool. The laminates were then sectioned into layers via careful milling, and a DSC scan was completed to assess crystallinity for each layer independently. Figures 3.3 and 3.4 show dramatic crystallinity variation through the thickness for the two laminates, at most plies far less than the maximum 26 – 29% PEKK crystallinity. In Figure 3.3 the laminate crystallinity elevates to 23.51% at ply 1 because it contacted the 175°C heated aluminum tool and was annealed (above its 154°C T_g) throughout the fabrication. Above ply 1, the crystallinity drops, beneath 2% for some layers.

In Figure 3.4, the crystallinity is even lower on the first ply at only 11.82%, as the tool is unheated. No mechanism to build crystallinity during placement is available other than the slow placement of ply 1 and the reheating of each of the 'n' plies as they are placed over. We are investigating this effect with alternate resins.

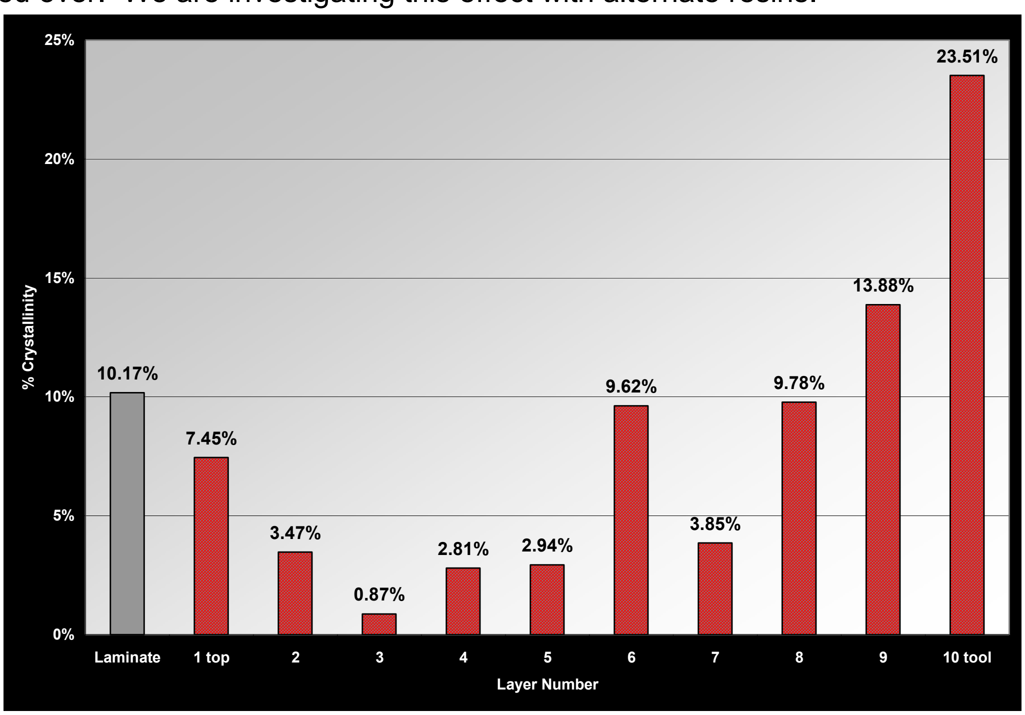

Figure 3.3 Crystallinity through the thickness for an AS-4/PEKK laminate processed on an aluminum tool heated to 175°C. Maximum PEKK crystallinity is 26% to 29%.

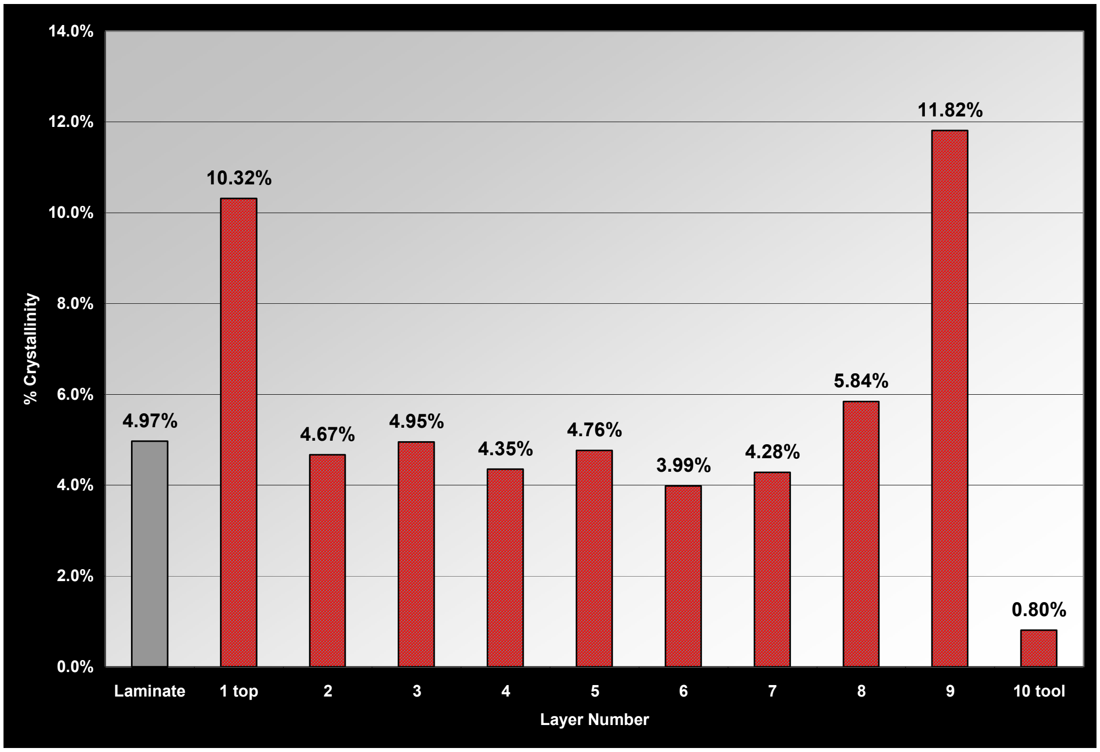

Figure 3.4 Crystallinity through the thickness for an AS-4/PEKK laminate processed on an unheated insulated composite tool.

Some initial data was also available for PEEK and PEKK tapes. PEEK tapes had 13.7% and 15.1% crystallinity out of a maximum expected of 34%-35%. PEKK tapes had a crystallinity of 2% and ~0%. It appears that the tape fabrication process also had cooling kinetics inadequate to build full crystallinity. The resin moduli for fully crystalline PEKK and PEEK are 3.4 GPa and 3.5 GPa, respectively. It has been estimated that the resin moduli can drop 30% if they are fully amorphous.

If this effect holds true, the thermoplastic community is in a conundrum. The most mature thermoplastic resin is semi-crystalline PEEK, yet its ability to build crystallinity out of the melt might not be sufficiently quick following in situ placement to build the full crystallinity required to manifest the resin modulus that supports the compression load-carrying capability and thus open hole compression strength. Recall that the time at temperature in the autoclave would be plenty long enough to build full crystallinity.

What can be done? Laminates could be placed more slowly than the current 6 mpm (20 fpm), but process economics would be jeopardized. The deposition head could be made longer to extend time at temperature, but the head would end up unwieldy, compromising the ability to place process features. Or the resin could be made to oven anneal following placement.

The best option would be to propose to the materials companies that a material system be designed for in situ tow and tape placement. This resin would need to be either fully amorphous (but keeping in mind the solvent resistance issues), be semi-crystalline with the ability to quickly crystallize commensurate with the in situ process cooling kinetics, or be amenable to post-placement-annealing to build crystallinity.

CONCLUSIONS

Voids cause loss of compression strength in cylinders and also play havoc with shear strength. Loss of shear strength acts synergistically with layer waviness to compromise compression strain-to-failure. In flat placed laminates, microstructure has been consistently observed that is void-free and absent from layer waviness. However, the time-temperature characteristics of the process have not allowed the full crystallinity of candidate polymers to be developed. This is under investigation with more semi-crystalline polymers like PEEK. Lack of crystallinity lowers resin modulus and compression properties suffer. We recommend the development of placement-grade tows and tapes designed specifically for the in situ process.

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