INVESTIGATIONS INTO LIGHTWEIGHT SOLUTIONS FOR IMPROVEMENT OF FIRE PROPERTIES OF EPOXY COMPOSITES

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Abstract

Aim is to improve the fire properties of epoxy composites with interlayers, without compromising the structural performance, i.e. by using a carbon fibre veil with expandable graphite, an intrinsically flame retardant thermoplastic film or a carbon fibre reinforced thermoplastic (CFTP) layer. Most promising seems the CFTP layer, while the mechanical performance appears unaffected.

1. Introduction

With more aerospace parts being made from polymer composite materials, many efforts are undertaken to improve the components' mechanical performance in the case of fire. Current solutions rely on fire-retardant additives in prepreg materials [1], though further improvement of the composite laminate can enable a prolonged structural stability. This study investigates three possible solutions for improvement of the fire safety, while protecting the structural properties of the polymer composite.

The first solution comes in the form of a fire retardant lightweight veil based on carbon fibres with expandable graphite on the laminate surface. In the case of fire, expansion of the veil should protect the underlying laminate. The second solution applies an intrinsically flame retardant high performance thermoplastic (TP) interlayer, which melts and expands during fire and provides an air barrier to delay the burning progression into the thickness of the laminate [2]. The third solution uses a thermoplastic layer including reinforcing fibres, such that this layer is also capable of contribution to the structural behaviour of the laminate, while providing a thermal barrier in the case of fire, leading to a multifunctional laminate.

The modified laminates with the veils at the surface and thermoplastic (composite) layers exchanged for various prepreg layers were investigated by means of cone calorimetry and residual compressive strength after impact to determine the optimal configuration in terms of fire resistance and mechanical properties. Investigations into laminate integrity during heat impact using the cone calorimeter were also performed. The quality of the manufactured panels was assessed using microscopy and ultrasonic C-scanning.

2. Experimental work

2.1. Materials

A 300mm wide and 127µm thick unidirectional (UD) prepreg tape Hexply® M21E with 34wt% resin content and IMA carbon fibres from Hexcel (F) was used. Ultem®1000 polyetherimide (PEI) film with 125µm thickness and 600mm width were acquired from Ajedium (USA). 75 mm wide HTS carbon fibre reinforced PEI UD tape (TPUD PEI-HTS40) was obtained from Toho Tenax (J) with a resin content of 34wt% and 175µm thickness. Various specimen sheets ~160mm in diameter of carbon fibre veil with expandable graphite (experimental Tecnofire® grades with 50wt% CF and 50wt% EG) were received from Technical Fibre Products (UK) with an areal density of 100±5g/m². Two types of EG were tested with activation temperatures of ~180°C and ~260°C (as determined with thermal gravimetric analysis), respectively.

2.2. Specimen preparation

Panels measuring 400x400mm² with a thickness of 2mm were manufactured for cone calorimetry testing. The specimens and their lay-up are presented in Table 1. It must be noted that the prepreg and TPUD had cuts in the single plies due to the smaller roll widths compared to the plate dimensions.

Specimen name	**Materials**	**Lay-up**
Reference (REF)	Prepreg	$[+/-/90/-/0/+/90/0]_S$
TP2-4	Prepreg, TP film	[+/TP/90/TP/0/+/90/0/0/90/+/0/-/90/-/+]
$TP2\text{-}4_S$	Prepreg, TP film	$[+/TP/90/TP/0/+/90/0]_S$
TPUD1	Prepreg, TPUD Tape	[(TPUD)/-/90/-/0/+/90/0/0/90/+/0/-/90/-/+]
TPUD2	Prepreg, TPUD Tape	[+/(TPUD)/90/-/0/+/90/0/0/90/+/0/-/90/-/+]
TPUD1-3	Prepreg, TPUD Tape	[(TPUD)/-/(TPUD)/-/0/+/90/0/0/90/+/0/-/90/-/+]
Veil180	Prepreg, CF+EG veil	$veil[+/-/90/-/0/+/90/0]_S$
Veil260	Prepreg, CF+EG veil	$veil[+/-/90/-/0/+/90/0]_S$

Table 1. Specimens and their lay-up for cone calorimeter testing (+:+45°, -: 135°).

Specimens for residual compression strength after impact (CAI) tests were performed only for selected specimens with TPUD interlayers, based on the cone calorimeter results. Panels of 400x400 mm² were manufactured according to the lay-up in Table 2.

Specimen name	**Materials**	**Lay-up**
Reference (REF)	Prepreg	$[+/0/-/90]_{4S}$
TPUD2	Prepreg, TPUD Tape	$[+/(TPUD)/-/90]/[+/0/-/90]_{3S}/[90/-/0/+]$
TPUD1-3	Prepreg, TPUD Tape	$[(TPUD)/0/(TPUD)/90]/[+/0/-/90]_{3S}/[90/-/0/+]$

Table 2. Specimens and their lay-up for CAI.

Specimens were manufactured using an Atlas autoclave with a vacuum build-up as shown in Figure 1.

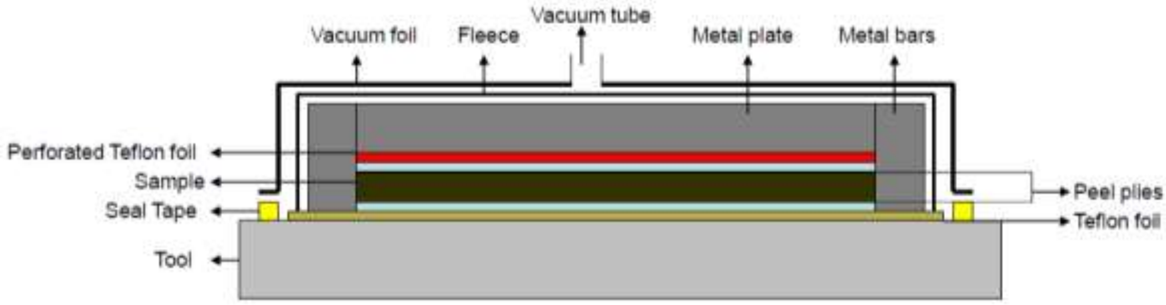

Figure 1. Vacuum build-up for autoclave curing.

The autoclave cure cycle consisted of heating to 180°C with a heating rate of ~2°C, holding for 120 minutes, and cooling to room temperature with 2°C/min under a pressure of 700kPa. The 2mm thick specimens for cone calorimeter were cut with a band saw into dimensions 100x100mm² and subsequently wrapped in aluminium foil (sides and bottom only) for the fire testing. The 4mm thick specimens for compression after impact testing were cut into specimens with dimensions 100x150mm².

2.3. Test set-up

A cone calorimeter from Fire Testing Technology Ltd. (UK) was used for assessing the materials' behaviour under high heat impacts according to ISO 5660. Two heating rates were applied: 35 and 50kW/m², which resulted in cone temperatures of 667 and 763°C, respectively. The distance between the specimen in the sample holder and the cone was 25mm. Due to the limited availability of the carbon fibre veils with expandable graphite, the specimens with these veils on the surface were tested only at 50kW/m². Per laminate and heating rate three specimens were tested, apart from the specimen with veil180, where only one specimen was available. Additionally, several specimens of each sample were tested in the cone calorimeter at 50kW/m² and stopped after different times between 30 and 120 seconds to study the effect of the heat impact over time.

Microscopy was performed with a Keyence Digital Microscope VHX-500 FD to assess the quality of the cross-section of the laminates after they were cut, embedded in resin, grinded and polished.

A Hilger USPC 3040 C-Scanning device with Hillgus® software was used to test the quality of the laminates for CAI in a non-destructive manner. In addition, this equipment was used to assess the damage area after impact of the specimens. This impact was performed for the CAI tests using a Myrenne Impact tower with an impactor of 15.75mm diameter and 2.49 kg weight, to enable a controlled impact with 10, 20, 30 and 40J. For each impact energy and laminate lay-up one specimen was tested.

The residual compression strength after impact tests were conducted according to AITM 1.0003 on a Zwick 1494 test (500kN) machine. The specimens were preloaded to 100N with 5mm/min and tested with 1mm/min.

3. Results and discussion

3.1. Laminate quality assessment

It was observed that due to the cuts in the TPUD plies, some resin of the prepreg had leaked through the junctions, see also Figure 2. This was most significant for the TPUD1 panel.

Figure 2. TPUD1, TPUD1-3 and TPUD2 panels after curing with peel-ply still on top.

Visual observations and micrographs of the laminates with the CF-EG veil showed that the veil was not always fully impregnated through the thickness with resin after curing, the effects of which remain to be investigated. Micrographs of the other laminates showed in general good consolidation and little porosity, though the CAI Specimen TPUD2 had some more porosity. The TPUD layers were found to be of somewhat larger thickness than the prepreg layers though this did not lead to a significant difference in thickness of the final plates. The C-scan results showed an effect of the resin flow at the TPUD junctions due to the increased local resin content.

3.2. Cone calorimeter

The main results of the cone calorimeter tests are summarised in Figure 3. Large scatter exists in the results, which is more apparent for time-related parameters, such as time to ignition and to peak-heat-release. As a general result, Figure 3b we see a large drop in peak release rate due to the addition of the modified layers, which is important for the final temperatures to develop in the case of a fire.

Visual inspection of the specimens after 120s at a heat impact of 50kW/m² learned that the back of the TP2-4, TPUD1 and TPUD2 specimens showed much less degradation of the composite than the reference, see Figure 4.

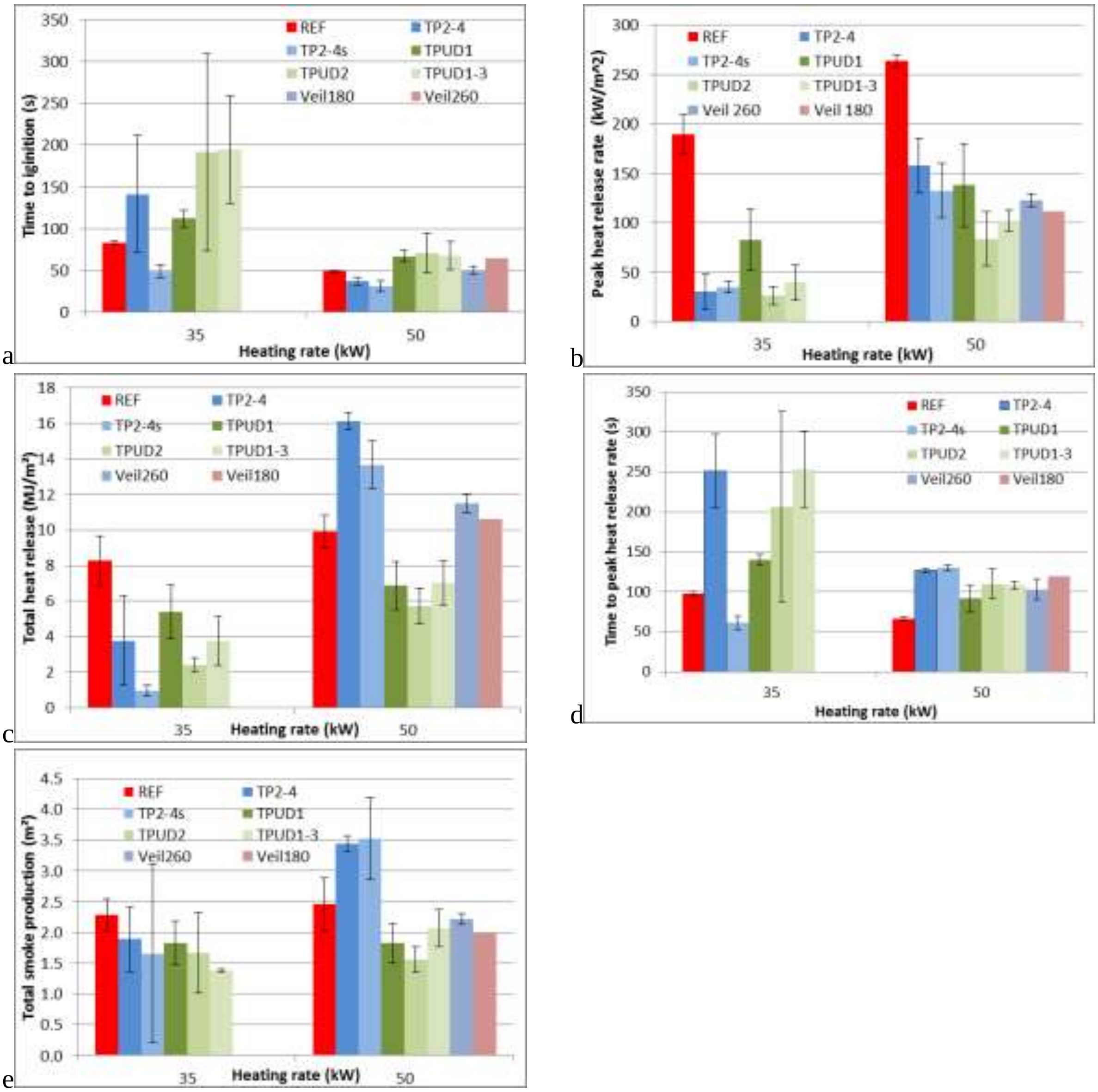

Figure 3. Overview of cone calorimeter results: a – time to ignition, b – peak heat release, c – total heat release, d – time to peak heat release, e – total smoke production.

	REF	TPUD1	TPUD2	PEI2-4
120s 50kW/m² back				

Figure 4. Photographs of specimen back surface after 50kW/m² heat impact for 120s.

3.2.1. TP interlayers

The effect of the incorporation of the TP interlayers is presented for the heat release rate (HRR) in Figure 5 for 50kW/m² heat impact. Two peaks can be seen for the TP modified specimens, which can be explained as follows: due to the heat impact on the surface, the TP layers just below the surface melt and expand leading to increased heat input in the surface. This also explains the shorter time-to-ignition, since due to the barrier effect of the molten and expanded TP layer, more heat and gases are collected at the surface leading to a locally higher temperature and earlier ignition, while protecting the layers underneath. This even leads to the fire extinction at a certain point due to the intrinsically fire retardant behaviour of this high performance thermoplastic. Upon continuous heat impact from the cone, the material starts to combust again when the first layers have been degraded.

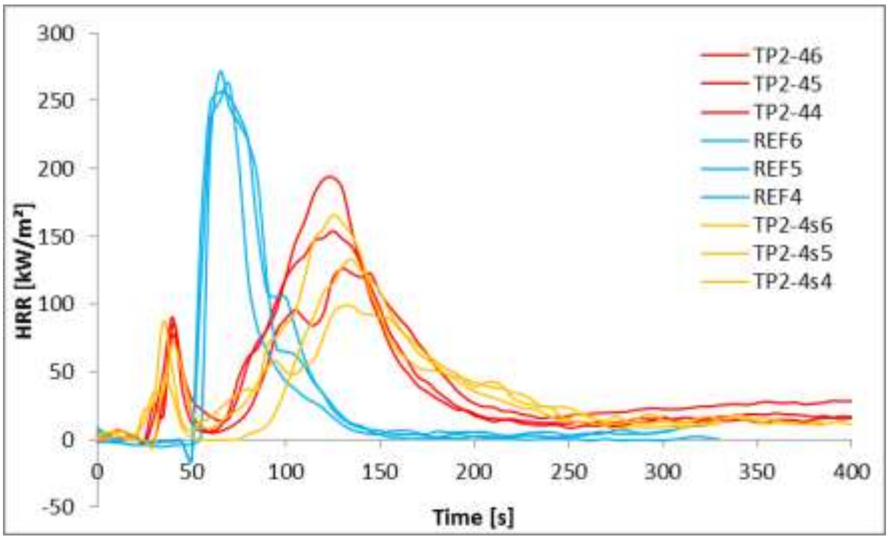

Figure 5. Heat release rate for reference samples and samples with TP interlayers at 50kW/m².

The peak heat release is significantly reduced and delayed by the incorporation of the TP interlayers compared to the reference material. For a heat impact at 35kW/m², the fire is even extinguished after burning of the surface and does not re-ignite again in contrary to the reference material, which does burn at this heat impact. This extinguishing behaviour for the TP modified specimens at 35kW/m² also explains the large scatter in results in Figure 3.

No significant difference was found for the lay-up of the TP interlayers, i.e. symmetrically or not. The only effect of the dimensional stability of the laminate, i.e. the non-symmetric configuration led to curved plates due to residual stresses [3].

3.2.2. TPUD interlayers

The results for specimens including TPUD interlayers in comparison to the reference material show a significant increase in time-to-ignition, especially at 35kW/m² heat impact; i.e. sometimes the material (TPUD2 and TPUD1-3) did not combust at all or only very little. The peak heat release rate (PHRR), total heat release and smoke production are significantly reduced while the time to reach the PHRR is much longer.

Of the various configurations, the specimens with the TPUD layer on the surface (TPUD1) seem to provide least benefit, which may also be explained by the 'leaking' of the prepreg thermoset resin through the TPUD tape joints. This is also shown by the heat release rate (HRR) results at 50kw/m² for the various specimens in Figure 6. Here, it can be seen that the effects of the TPUD2 and TPUD1-3 lay-up on the HRR are comparable, though the TPUD1-3 seems to provide more reproducible results.

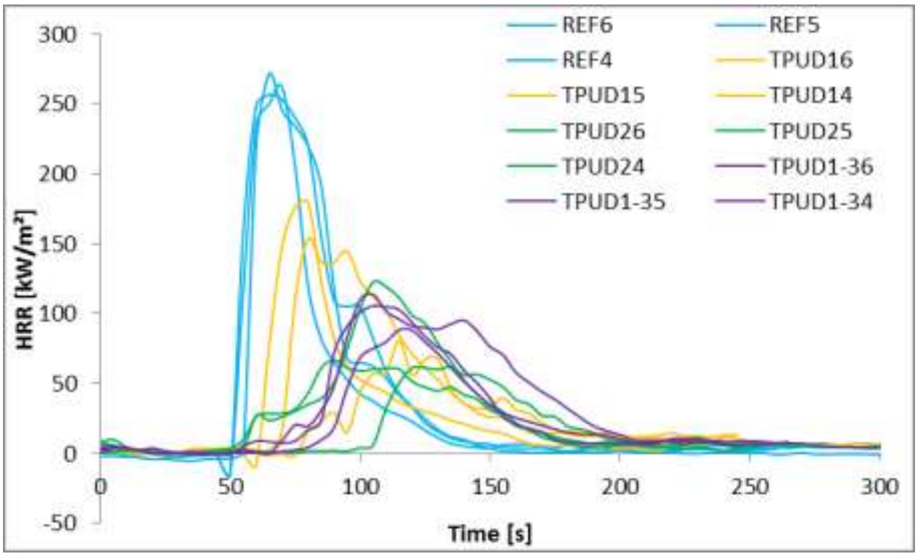

Figure 6. Heat release rate for reference samples and samples with TPUD interlayers at 50 kW/m².

3.2.3. Carbon fibre veils with expandable graphite

The specimens with the carbon fibre veils with expandable graphite show generally a delay in combustion behaviour, such as smoke production, heat release and mass loss. Also the peak heat release is much less and occurs later than for the reference material due to the barrier properties of the flame-retardant veil. An example of the results for the veil-modified specimens compared to the reference material is shown in Figure 7.

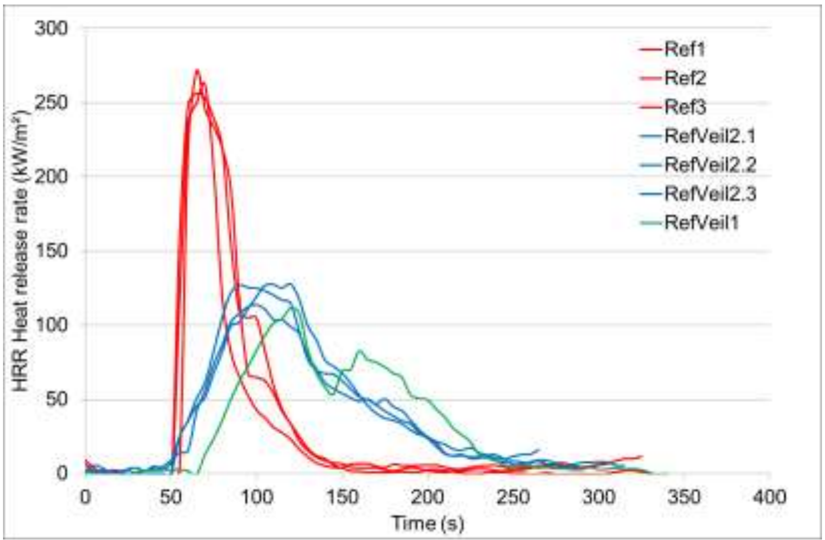

Figure 7. Heat release rate at 50kW/m² for reference samples and samples with the CFEG veils (veil1 = veil180, veil 2 = veil260).

The most effective modification as determined with cone calorimetry seems to be the addition of TPUD layers when looking at the critical parameters time-to-ignition and peak heat release rate: the longer times and lower peak results in more time for evacuation of the passengers in case of fire and less heat development. Hence, it was decided to perform mechanical tests with these specimens to check the effect on the mechanical properties.

3.3. Mechanical testing

The compression after impact results are shown in Figure 8.

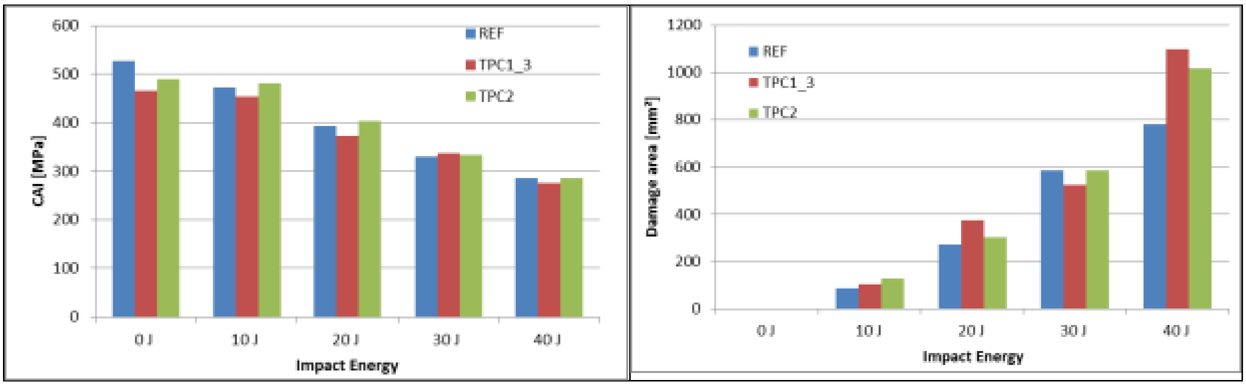

Figure 8. Compression strength (left) and damage area (right) of the laminates for several impact energies.

As expected, the residual strength after impact was found to decrease with increasing impact energy for all specimens, as well as the increase of damage area. Based on results and the limited number of tested specimens, no significant difference can be observed in CAI results between the different specimens, which may indicate that the improvement of combustion behaviour by adding TPUD layers does not compromise significantly the structural performance of the material.

4. Conclusions

The addition of surface and interlayers of various consistencies (carbon fibre veil with expandable graphite, intrinsically flame retardant thermoplastic film –TP- and this film reinforced with carbon fibres -TPUD) and various lay-ups seem to improve the combustion behaviour of epoxy prepreg composites as determined by cone calorimetry. Especially at low heat impacts (i.e. 35kW/m²) the material can be extinguished. The TPUD modified specimens seem to provide most benefit and the CAI properties were not found to be affected significantly. Future work will consist of deeper investigations into the burning behaviour and more mechanical testing of these modified laminates.

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