

THE CASE FOR MACROSCALE THROUGH-THICKNESS REINFORCEMENT FOR HYBRIDISED MULTIFUNCTIONAL COMPOSITES

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ABSTRACT

Embedding fundamental functional properties like thermal and electrical conductivity in composites is key in deriving a range of additional industrially-useful functionalities. Through-thickness reinforcement (TTR) can achieve this via material hybridisation. These techniques are sometimes unattractive due to the need for specialised equipment and a shortage of available expertise to support TTR. A knowledge gap exists in understanding the interplay between reinforcement size and functional behaviour especially where reinforcements go beyond traditional geometrical constraints. Here we demonstrate that the addition of macroscale (≥ 1 mm) TTR can significantly improve electro-thermal energy transfer as well as provide effective tailoring functionality in composites.

1. INTRODUCTION

Fibre reinforced polymer composites are seen as a key enabling technology in realising Net Zero. Functional through-thickness reinforcement (TTR) in composites is a growing area of research interest and industrial demand aligning with the strong uptake of high-performance composite materials outside of aerospace [1]. TTR is primarily used to improve delamination resistance [2] or in joining applications [3] but have substantial practical implications for the integration of functional elements for thermal/electrical management [4,5] or sensing [6]. From these fundamental behaviours, a series of addition functionalities can be derived like EMI shielding, self-healing, and etc. Existing TTR methods like tufting, stitching, and z-pinning place strict limitations on the reinforcement's elasto-plastic and geometric properties [7] and require specialist equipment and expertise that is currently scarce across the European composites base. Considering (quasi-)static insertion of TTR vastly expands the material and geometric configurations possible and is compatible with existing manufacturing routes, like hot presses, which are in greater use outside of aerospace.

Z-pinning includes the use of ultrasonication to insert submillimetre (0.2-1.0 mm) diameter pins and is generally more suitable for prepreg manufacturing routes [8]. Pins are typically metallic, polymeric, or composite, to achieve the desired stiffness need for insertion, and are applied in low vol.% (0.5 – 4 vol.%) arrays. Typical in-plane knock-down of elastic properties is dependent on pin diameter and vol% and is between 10-15% but is accompanied by significant improvements in interlaminar properties by up to an order of magnitude [9]. It is worth noting that the majority of elastic property related studies on z-pinned laminates have been performed on unidirectional (UD) laminates in line with more traditional aerospace interests. There are also well-reported complications in terms of keeping the pins perfectly

oriented in the through-thickness direction, especially as preform thickness increases. The hybridisation of composites with conductive z-pins has been proven to result in increases in through-thickness thermal conductivity by up to ~650% [10] and electrical conductivity by up to six orders of magnitude [11] by using metallic or carbon-based pins.

Static pin insertion is more suitable for larger (1 – 2 mm in diameter), rigid pins [12] and is compatible with both prepreg and dry fibre routes with no ultrasonication needed. A lower chance of bucking and low insertion forces [12] means that this process can be directly integrated into typical stamp or hot-press forming and curing methodologies. The use of this technique in UD laminates is not well explored and is likely to cause significant mechanical property degradation, however in woven laminates the local effect of the pins on tensile strength has been reported as insignificant, with up to a doubling in stiffness but a significant (up to ~50% loss) in compressive strength being noted [13]. These large diameter pins have also been proven to locally affect the thermal energy transfer in the material around the pin [14] and having different pin-laminate heat transfer interactions than z-pinned laminates [15].

On the whole, mechanical response in pinned laminates is heavily governed by the laminate mesostructure, pin configuration (including density, pattern, insertion depth) and the fiber bed topology. Given that these planar knock-downs are more acceptable outside aerospace, there is a need to better understand how material hybridisation via pinning can be exploited to embed functionality into structures alongside the optimisation of this operational envelope for the desired application in line with manufacturing and weight considerations. The challenge is that submillimetre scales of TTR in composites require specialist equipment/expertise and adds and additional manufacturing steps to component fabrication. Although z-pinning has proven capable embedding functionality, can macroscale (≥ 1 mm) pins provide similar functional improvements alongside more convenient integration? Here we demonstrate that macroscale z-direction TTR can improve three-dimensional thermal and electrical energy transfer as well as provide effective tailoring of this functionality in composites.

The novelty of this work is in demonstrating that macroscale (≥ 1 mm) TTR can be used to achieve noticeable improvements in electro-thermal energy transfer and that the mechanisms by which the TTR facilitates electro-thermal energy transfer is not directly scalable (e.g. by rule of mixtures). Instead, this is a complex interplay between the synergistic effect between the pins and the pin-laminate interface. The research contributes to addressing the demand for TTR methods capable of embedding functionality using larger functional elements, which in turn reduce manufacturing complexity and time, not currently possible with established TTR methods. This will have significant impact by supporting the drive towards embedding multi-functionality in composites alongside positive implications for sustainability through repair and recycling.

2. METHDOLOGY

2.1 Experimental

The fibre reinforced composite material systems used in this work were the 2 × 2 twill 12k (650gsm) and 3k (200gsm) variants of BX180-220 carbon-benzoxazine supplied as a prepreg and manufactured by SHD Composite Materials Ltd. For experimental validation, TT pins were manufactured from 2.0 mm diameter 304L stainless steel rods. Through-thickness pin insertion was carried out using a bespoke insertion gun in accordance with the procedure in [12]. The laminate consisted of 15 layers [(0/90F,±45F)₇,0/90F] of 12k and 1layer of 3k [±45F]

prepreg material (16 layers total) with a measured average thickness in the pinned zone of 8.0 ± 0.6 mm. The prepreg stack was consolidated under vacuum pressure at 30 °C and afterwards was further heated and maintained at ~ 60 °C to allow for in static pin insertion. For the validation coupon, the pin pitch was 10 mm and pins were arranged in a 3×3 configuration shown in Figure 1a. The stack with inserted pins was cured at 180 °C under 6 bar autoclave pressure in accordance with the manufacturer's specifications.

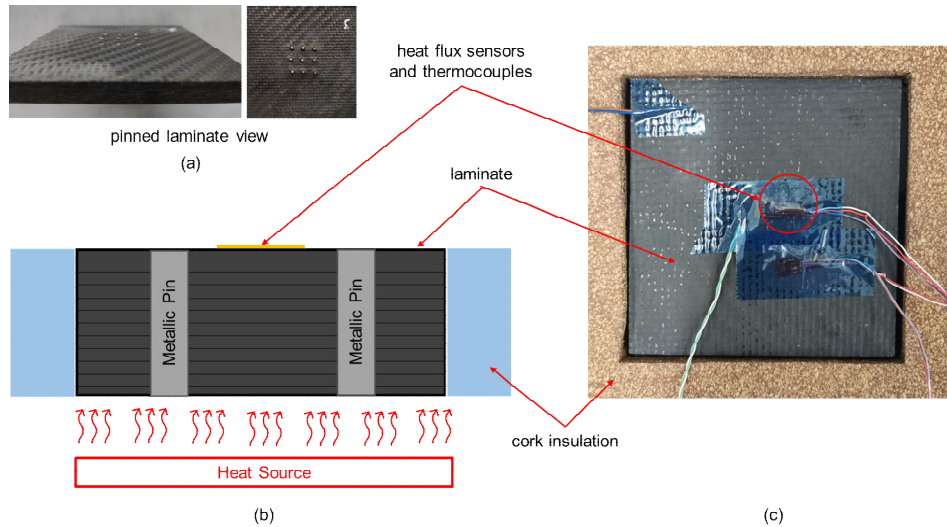


Figure 1: Experimental setup of steady state heat transfer experiments showing manufactured pinned laminates, (b) through-the-thickness schematic, and (c) experimental configuration.

This work uses the BX180 laminate (experimental and FE simulation), 304L stainless steel pins (experimental and FE simulation) and copper pins (FE simulation). Their thermal and electrical properties are given in Table 1. Composite properties are experimentally derived in [14,16], pin properties are taken from the Granta Selector material database.

Table 1: Material Characterisation data input for FE simulation.

	Specific Heat capacity (J/(kg·K))	Density (kg/m ³)	Thermal Conductivity (W/(m·K))	Electrical Conductivity (S/m)
Stainless steel (AISI 304)	471 (11 °C) to 501 (215 °C)	8010	14.9 (21 °C) to 18 (204 °C)	1.45e6 (-15 °C) to 1.15e6 (228 °C)
Copper (C10100)	383 (16 °C) to 413 (303 °C)	8940	397 (14 °C) to 387 (227 °C)	5.64e7 (10 °C) to 3.48e7 (199 °C)
BX180-220	943 (25 °C) to 1702 (180 °C)	1584	K ₁₌₂ 3.3 (25 °C) to 5.1 (180 °C) K ₃ 0.81 (25 °C) to 1.25 (180 °C)	1.75e4 (planar) 40 (out-of-plane)

*note full temperature dependent tables are used in FE simulation but only ranges are given here for conciseness.

To validate the FE model and to determine suitable boundary conditions, steady state 1D heat transfer experiments were performed (Figure 1b and c). The laminate panel was placed on a 600 × 600 mm ELKOM RTM heating table that provides very uniform heat flux across its surface (maximum temperature variation measured at ± 1.25 °C) and was insulated around its edges with a 10 mm thick cork rim with thermal conductivity of 0.12 W/m·K and the gaps between the cork and the laminate were plugged with breather fabric (not shown). A thin layer

of thermal grease was applied at the interface between the composite panel and the hot plate to improve thermal contact and the exposed surface of the composite panel was sprayed with Graphit 33 graphite spray to reduce radiation losses and improve visualisation from the thermal camera. Thermocouples and heat flux sensors were then applied to the composite surface (Figure 1c), to monitor top surface heat transmission alongside the thermal camera.

2.2 Finite Element Model

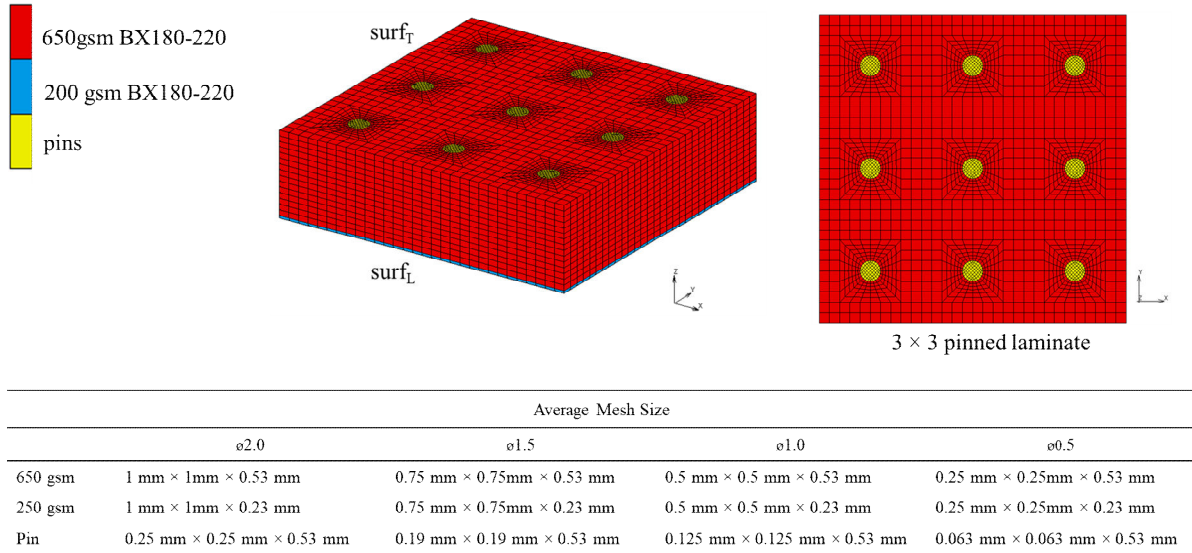


Figure 2: RVE FE model showing geometry and mesh used for analysis.

A mesoscale (ply-level) current/thermal finite element simulation consisting of a 3 × 3 pin representative volume element (RVE) was developed in the Marc [17] finite element software and is shown in Figure 2. The RVEs consisted of 2 main components – the laminate and the pin. The laminate is modelled as 16 layers of full integration hex continuum elements (element number 43 in Marc notation), each corresponding to a single layer of material. The metallic pins are also modelled similarly for congruence with the laminate mesh and element refinement in the region of the pin. Details of the FE model is given in Figure 2 including average element sizes and distributions which scale with respect to the pin size and laminate thicknesses evaluated. The pin volume fraction (vol%) is constant throughout all simulations at 3.1% with pin decreasing from 10 mm in $\phi 2.0$ pinned laminates to 2.5 mm in $\phi 0.5$ pinned laminates. Laminate orientations are defined in accordance with the layup scheme given in Section 2.1 and is assigned an orientation of (1 0 0) for 0/90° plies and (1 1 0) for $\pm 45^\circ$ plies. Thermal periodic boundary conditions (PBCs) have been enabled to give the spatial repeatability. PBCs are formulated as shown in equation 1. where u_t represents the degree of freedom to be constrained, $u_1, \dots, u_n$ are other retained degrees of freedom of the structure, and $a_1, \dots, a_n$ are constants.

$$u_t = a_1 u_1 + a_2 u_2 + \dots a_n u_n \quad (1)$$

In Marc, PBCs are implemented using servo-links which prescribe multi-point boundary conditions for nodal displacements expressed as a linear function with constant coefficients. Control nodes are created for the x and y faces next to the negative faces of these directions. Then the nodes on the negative face were selected as the constrained nodes and the nodes on

the positive face and the control node are set as the retained nodes with a coefficient of 1 for the positive face and coefficient of -1 for the control node. This results in the modification of equation 1 to equation 2 where u_t corresponds to the DOFs of at the negative face, u_1 refers to DOFs at the control node and u_2 corresponds to the DOFs at the positive face. The external boundary condition at the control nodes (and hence RVE sides) is set to a zero-flux and zero-potential condition.

$$u_t = -1u_1 + 1u_2 \quad (2)$$

Within the FE model, it is important to note the following assumptions and simplifications (i) ideal contact between composite and heat source was assumed; (ii) local tow distortion caused by the pins was ignored; (iii) constant thickness and parallelism between the top and bottom faces was assumed.

2.3 Finite Element Loadcases

The finite element boundary conditions and loadcases are discussed more thoroughly throughout the results section at the points where they become relevant to the understanding of the results text. However, Table 2 gives an overview of the loadcases examined and how they relate to different sections of the results.

Table 2: Thermal simulation loadcases.

Loadcase	Temperature (°C)	Heat Flux (W/m ²)	Film Coefficient (W/m ²)	Current Density (A/m ²)	Voltage (V)	Relevant section
LC1	75 (surf _L)	-	12.1 (surf _T)	-	-	3.1
LC2	25 (surf _L + surf _T)	3×10 ⁷ (surf _L)	25 (surf _L + surf _T)	-	-	3.2
LC3	75 (surf _L + surf _T)	3×10 ⁷ (surf _L)	75 (surf _L + surf _T)	-	-	3.2
LC4	150 (surf _L + surf _T)	3×10 ⁷ (surf _L)	150 (surf _L + surf _T)	-	-	3.2
LC5	25 (surf _T)	-	25 (surf _T)	A: 2e5 B: 3600 C: 400	0 (surf _T)	3.3

*surf_T = top surface and surf_L = lower surface

Zone 1A lightning strike test simulations followed standard MIL-STD-1757A, comprising three current components: the fast waveform A and the slow waveforms B and C.

3. RESULTS

3.1 Model Validation

Figure 3 shows the comparison between temperature profile between the FE model and validation experiment in laminates pinned with 2 mm diameter stainless steel pins. The steady state lower surface (surf_L) temperature is 75±0.2 °C, the average measured top surface (surf_T) temperature is 65±0.2 °C (from thermocouples), and the heat flux at surf_T is 497±24W/m² (from heat flux sensors). The loadcase used in the simulation is LC1 according to Table 2. The film coefficient (h) for convection losses through the air was derived by combining Fourier's law

and the principle of energy conservation into equation 3, using the experimental heat flux values measured at the exposed surf_T in the region of the pins.

$$-k \frac{dT}{dx} = h (T - T_{\infty}) \quad (3)$$

Where k is temperature dependent thermal conductivity, dT/dx is the temperature gradient, h is the film coefficient, T is the surface temperature and T_{∞} is the environmental air temperature ($T_{\infty} = 21$ °C). By applying the steady state heat flux and temperature values obtained, film coefficient is calculated to be $h = 12.1$ W/m², which agrees with the range of h values for non-forced convection of air of $h = 2.5 - 25$. The FE simulation shows surf_T temperature is 70 °C, and the heat flux at surf_T is recorded as 586 W/m².

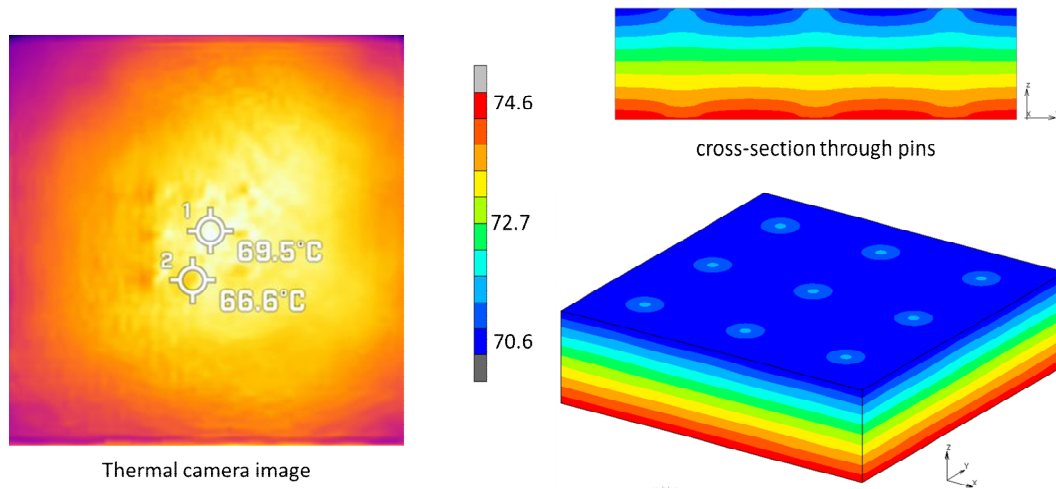


Figure 3: Model validation comparing experimental with simulative results.

There is generally good agreement between the simulation and experimental results with a 7.4% and 16.4% percentage disparity between experimental and simulation values for temperature and heat flux at surf_T respectively. The much-increased simulative heat flux suggests this is likely due to the assumption of perfect thermal contact between the laminate and heat source as well as the assumed zero heat losses at the RVE edge. To increase confidence in the model, the experimental laser flash analysis (LFA) derived diffusivity results presented in [14] is compared with simulation derived values in greater detail in Section 3.2.

3.2 Effect of copper pin diameter on through-thickness thermal energy transfer

A heat pulse was applied to 8 mm thickness RVE to simulate a high energy heat pulse as is done in LFA. This is repeated for environmental conditions at 25 °C, 75 °C and 150 °C, representing the full range of the composites working temperature (T_g peak is at 180 °C). These environmental condition inputs to the model coincide with LC2 - LC4 in Table 2. A boundary condition was set for a heat pulse on surf_L of RVE model with a time length of 10^{-4} s and a heat flux of 3×10^7 W/m². By considering interfacial heat exchange in the FE simulation the time-dependent temperature change was obtained, and the simulated z-direction thermal diffusivity (α) was calculated according to Equation 3 from ASTM E1461-13R22.

$$\alpha = \frac{0.139L^2}{t_{1/2}} \quad (4)$$

Where α is thermal diffusivity, L is the laminate thickness and $t_{1/2}$ is the period when the detected temperature reaches half of the maximum. Thermal conductivity is then calculated according to equation 5.

$$K = \rho \cdot C_p \cdot \alpha \quad (5)$$

Where K is thermal conductivity, ρ is density and α is thermal diffusivity.

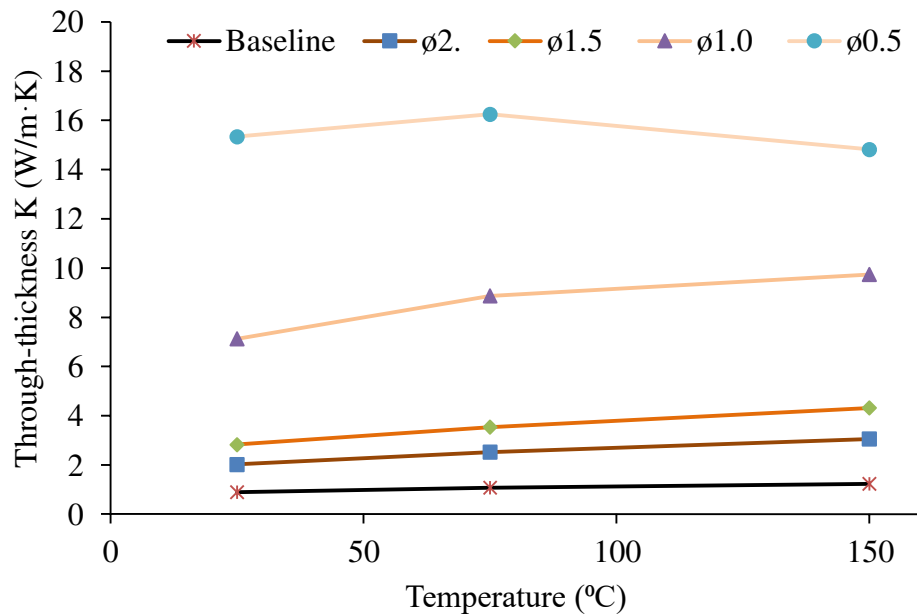


Figure 4: Through-thickness thermal conductivity of unpinned and pinned laminates.

Figure 4 shows the relationship between temperature and through-thickness thermal conductivity in unpinned and copper pinned samples derived from FE simulation results. Baseline thermal conductivity values obtained from the simulation agree well with experimentally derived inputs in Table 1. The presence of copper pins greatly increases the thermal conductivity in pinned samples by providing conductive paths that allow heat to flow in the through-thickness direction. Considering room temperature values, conductivity increases by 126% from 0.9 W/m·K in baseline to 2.0 W/m·K in ø2.0 mm pinned samples. The maximum increase on the baseline is in ø0.5 mm pinned samples in which thermal conductivity increases by 1619% to a maximum value of 15.3 W/m·K.

It is clear that, providing that the vol% of the pins remains constant, decreasing pin diameter (and hence spacing) has the effect of improving through-thickness heat conduction. The difference is due to the change in heat transfer mechanisms where there is a greater synergistic effect between the pins and between the pin-laminate interfaces that results more effective bulk heat transfer as is demonstrated in Figure 5. The comparison shows that there is more rapid dispersal of heat in ø0.5 pinned laminates where heat is dissipated not only up through the pins, but across laminate layers with high levels of exchange between the pin and the laminate. This is the synergistic effect of pins working together in contrast to ø2.0 pinned laminates where most of the heat is absorbed into the pin which acts as local heat sinks carrying the burden for most of heat transfer. This suggest that for TTR laminates, the efficiency of thermal energy

transfer cannot be estimated by simple analytical methods like rule of mixtures and is instead a function of pin diameter (likely the interfaces) and spacing.

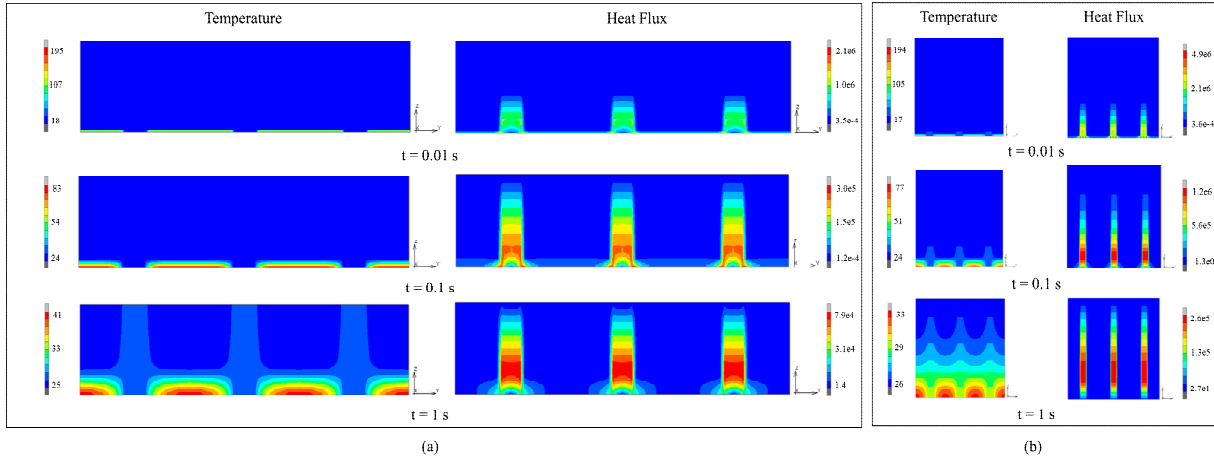


Figure 5: Contour plots at RVE cross-section showing laser flash pulse transmission in (a) ø2.0 mm copper pinned, and (b) ø0.5 mm copper pinned samples.

3.3 Three-dimensional electro-thermal energy transfer

The 3D electro-thermal behaviour of the ø2.0 and ø0.5 copper pinned laminates was investigated and evaluated. The laminate was subjected to a simulated zone 1A lightning strike test using the parameters in LC5 from Table 1 following standard MIL-STD-1757A with a maximum current in the A waveform peak of 50kA for the ø2.0 copper pinned laminates and 12.5kA for the ø0.5 copper pinned laminates (applied current density is constant in line with LC5).

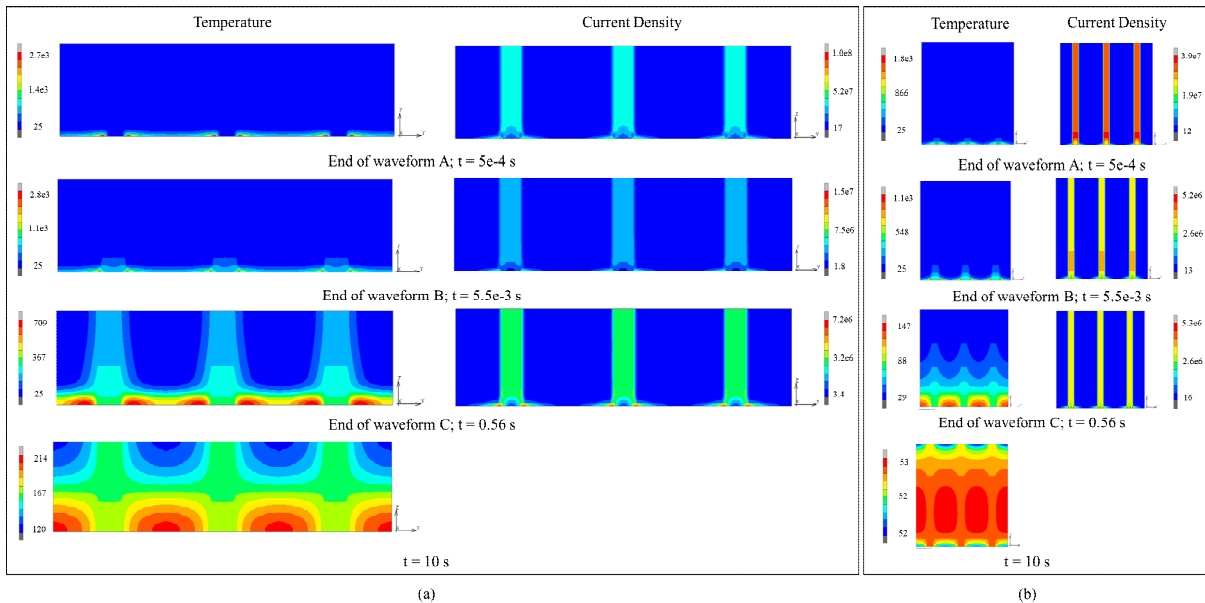


Figure 6: Contour plots at RVE cross-section showing zone 1A lightning strike energy transmission in (a) ø2.0 mm copper pinned, and (b) ø0.5 mm copper pinned samples.

Figure 6 shows the temperature-current evolution with time. Heat transfer through the laminate agrees with what is seen in the LFA simulations, where heat builds up at surf_L as temperature increases due to joule heating caused by the electrical input and is then transferred through the

laminate. As expected, based on the thermal conductivity results from Figure 4, $\phi 0.5$ mm pinned samples are able to dissipate heat more effectively resulting in lower temperatures throughout ($t = 0 - 0.56$ s) and after the strike ($t = 10$ s). In both pin configurations, large temperature concentrations are noted at the pin-laminate interface noticeable at waveform A, B and C (Figure 6). This is due to the large current influx from the laminate into the pin at the surface. As the local thermal gradient within the pin changes, so too does the temperature-dependent electrical resistivity gradient within the pin. This leads to a time-temperature dependent variation in the current flow vectors which changes the temperature profile at the interface throughout waveforms A and B. These large local temperature spikes could potentially be responsible for local pin-laminate interface failure due to degradation and/or thermal expansion mismatches. Another interesting line of future study could focus more locally on the Lorenz forces caused by the large current in the pins and generated electromagnetic forces which acting on the fibres could be a source of further afield delamination.

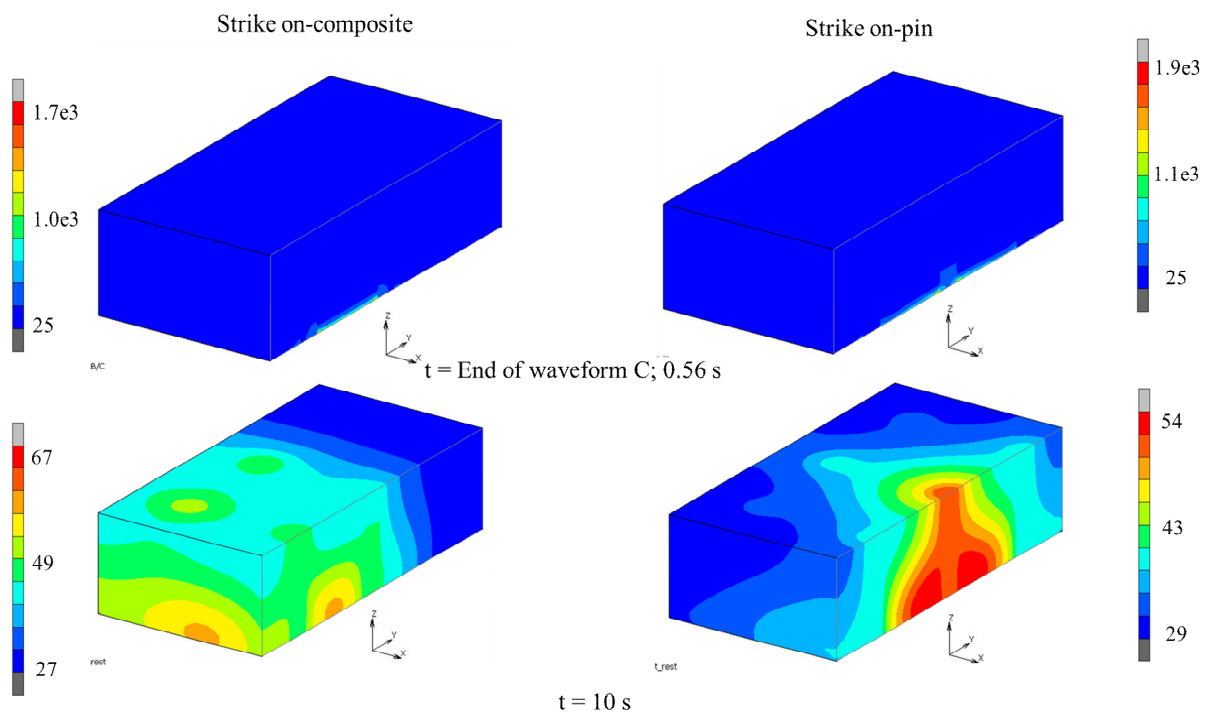


Figure 7: Contour plots at RVE cross-section showing zone 1A lightning strike thermal after effects of a 100mm^2 strike on the $\phi 2.0$ mm copper pinned laminate between the pins ("strike on-composite") or at the pin ("strike on-pin").

Figure 7 compares $\phi 2.0$ mm pins and comparing a local 10×10 mm strike at the space between the pins or directly at the pin with maximum A waveform current of 5kA (applied current density constant in line with LC5). Results show much improved heat transfer through the laminate thickness as opposed to pooling on the laminate strike surface in the event of a direct pin strike. This suggests that strike size and placement is integral in developing effective and efficient electricity and heat paths in the laminate by the use of TTR.

4. CONCLUSIONS

The work develops uses validated finite element model for a representative volume element of composite reinforced through-the-thickness with stainless steel and copper metallic pins of varying diameter between 0.5 and 2.0 mm. FE simulation was carried out using the Marc FE

solver and results for both pinned and unpinned laminates agree well with experimental characterisation results for steady state heat transfer and thermal conductivity.

The presence of highly conductive copper pins of all diameters between 0.5 mm and 2.0 mm improves through-thickness heat transfer in the pinned laminate compared to baseline material as the pins act as a thermally conductive pathways that accelerate the movement of heat energy through the material. Furthermore, it was shown that the pin diameter of the TTR has a significant effect on the efficiency of this heat transfer process. Larger pin diameters tend to conduct heat as independent heat paths rather than working in synergy between the pins and the laminate interfaces. TTR pins are shown to provide between a 126% and 1619% improvement in through-thickness thermal conductivity depending on the configuration. 1 mm diameter pins which are at the upper limit of z-pinning techniques but at the lower limit of static insertion techniques can provide significant improvements without the need for embedding of pins using specialist techniques. These morphologies can for example be manufactured on surfaces and driven into laminates. Thermal results suggest that for TTR laminates, the efficiency of thermal energy transfer cannot be estimated by simple analytical methods like rule of mixtures and is instead a function of pin diameter and spacing.

The electro-thermal behaviour of the pinned laminates was also investigated and concluded that TTR results in improved heat and electrical energy flow and is similarly a function of pin diameter and spacing. Interesting observations at the pin-laminate interface showed that additional heat generation at the pin-laminate interface is the result of large amounts of current flowing into the pin from the laminate and shows strong correlation to the material's temperature dependence. This warrants further study in future.

The novelty of this work is in demonstrating that macroscale (≥ 1 mm) TTR can be used to achieve noticeable improvements in electro-thermal energy transfer and that the mechanisms by which the TTR facilitates electro-thermal energy transfer is not directly scalable (e.g. by rule of mixtures). Instead, this is a complex interplay between the synergistic effect between the pins and the pin-laminate interface. The research contributes to addressing the demand for TTR methods capable of embedding functionality using larger functional elements, which in turn reduce manufacturing complexity and time, not currently possible with established TTR methods.

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