Experimental characterisation for compression moulding of hybrid architecture composites using reclaimed prepreg manufacturing waste

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Abstract. Composites manufacturing using prepreg general a large proportion of waste from ply cutting, which usually ends up in landfill. A novel reuse route for prepreg manufacturing waste is proposed by combining chip-SMC reprocessed from the waste material and the virgin continuous fibre prepreg to create hybrid architecture composites. Experimental studies are performed to investigate the flow behaviour of prepreg chip-SMC under typical compression moulding conditions and benchmark it against a conventional SMC. Process characterisation is also performed for hybrid architecture composites to understand the critical deformation mechanisms of chip-SMC and prepreg, and the interaction between the two materials. Compression moulding trials are performed to further study the material behaviour and process characterises under realistic manufacturing conditions.

Introduction

Composites manufacturing using prepreg typically generates 30%-50% of waste from ply cutting [1], which usually ends up in landfill, having negative impact on the environment and resulting in substantial costs. Research has shown that the CO_2 emissions associated with manufacturing virgin material is significantly higher (up to 80% [2]) than for retrieving fibres from manufacturing scrap. Developing commercially viable solutions to reclaim and reuse carbon fibre prepreg is therefore of great industrial interest, increasing material efficiency, reducing manufacturing cost, and most importantly avoiding landfill. The key to prepreg reuse is to handle the manufacturing waste “here-and-now” to eliminate further cost and emissions during transportation, developing intermediate products that fit readily within existing manufacturing processes and infrastructures.

Prepreg off-cuts typically contain fibres of a finite length, therefore any potential recycling strategy should focus on improving the processibility of this material to offset the disadvantages of the resulting discontinuous fibres, using intelligent topological design to maximise the mechanical performance. Several approaches have been explored for processing discontinuous prepreg, including chip-based sheet moulding compounds (SMC), which flow under heat and pressure to fill the mould cavity when compression moulded. Commercial examples using virgin prepreg include Hexcel’s HexMC and DowAksa’s Engineered Moulding Compound (EMC). This approach potentially offers the most robust way of reprocessing prepreg scraps, tolerating any size or shape of material off-cuts, utilising 100% of the waste. Existing prepreg-based SMCs usually consist of fibre lengths similar to conventional SMCs (e.g. 12.5mm, 25mm and 50mm), but the level of in-cavity flow is limited by the higher fibre content (>60% Vf compared to <40% Vf for conventional SMCs). Little effort has been made to investigate ways to engineer the flow characteristics of these materials, particularly to maximise the out of plane flow behaviour to fill complex geometries.

This paper proposes a practical solution for utilising prepreg waste, by co-moulding continuous fibre prepreg with discontinuous prepreg chips (SMC) to form composites with a hybrid architecture . A series of experimental studies have been performed to characterise the main deformation mechanisms for prepreg chip-SMC and hybrid architecture materials under typical compression moulding conditions. A bespoke squeeze flow rig has been used in this paper for all of the experimental characterisation studies. Compression mouldings of hybrid architecture composites combining virgin prepreg and chip-SMC have been conducted using a flat plaque geometry.

Material Characterisation Methodologies

Sample Preparation. The majority of the studies in this paper have been conducted using Mitsubishi TRK501 - an epoxy based woven carbon prepreg with 12k tow size, 2x2 twill weave and 55% fibre volume fraction. Due to the interruption to material supply, an alternative carbon/epoxy supplied by DowAksa with the same tow size, weave pattern and fibre volume fraction was used for characterising the pure chip-SMC specimens. The DowAksa material is not commercially available, therefore further details of the material will not be disclosed due to confidentiality reasons . All chips were cut at 0/90° in relation to the direction of the fibres, and three different chip sizes were studied, including 12.5mm, 17.2mm, and 25mm. Lamination of loose prepreg chips was carried out by shuffling the chips within a sealed plastic envelope until the distribution of the chips was visually uniform. Envelopes containing the chips were then positioned on the conveyor belt of a Schmidt & Heinzmann compounding line, and compacted at 3 bar and 50°C inside the compaction chamber of the compounding line. **Fig. 1** shows one sheet of chip-SMC consisting of 25mm square chips.

Fig. 1: A sheet of manufactured chip-SMC consisting of 25mm square chips

Chip-SMC Characterisation. Flow characterisation for chip-SMCs was performed on a squeeze flow rig shown in **Fig. 2**, which was originally developed in [3] for characterising the flow behaviour for conventional SMCs. The squeeze flow rig consisted of a pair of 350mm × 350mm testing plates heated by cartridge heaters. The testing plates were attached to a die set with four guide columns to ensure parallelism of the testing plates. An extensometer was attached to the side of the testing plates for accurately measuring the displacement of the moving plate. Circular-shaped squeeze flow testing specimens with a diameter of 100 mm (**Fig. 3** (a)) were stamped out from the laminated chip-SMCs using a clicker-press. Chip-SMCs of three different nominal areal masses were tested in this study: 3000gsm, 6000gsm and 8000gsm. All tests were performed at

100°C mould temperature, which was lower than the recommended moulding temperature of 150°C in order to slow down the curing of the material and provide the operator sufficient time to load the specimen and start the test. This temperature (100°C) was selected to ensure the resin viscosity remained constant. All tests were performed at a constant speed of 1 mm/s with a 200 kN force limit. At the end of the test, specimens were held between the plates at a constant cavity height for 20 minutes to develop a sufficient degree of cure before removing **Fig. 3** (b)).

The flow behaviour of each specimen was assessed in terms of the compressive stress-strain response and the area increase of the specimen after the squeeze flow testing. Detailed methods for determining the stress-strain response and the area increase of the specimen can be found in [3]. VORAFUSE® EMC supplied by DowAksa was selected as a baseline SMC in this study to benchmark the flow behaviour of chip-SMCs against commercial SMCs. The baseline SMC was a UD prepreg-based SMC consisting of the same epoxy resin system as the prepreg, with 27mm – 54mm fibre length, 1048gsm areal mass and 55% fibre volume fraction.

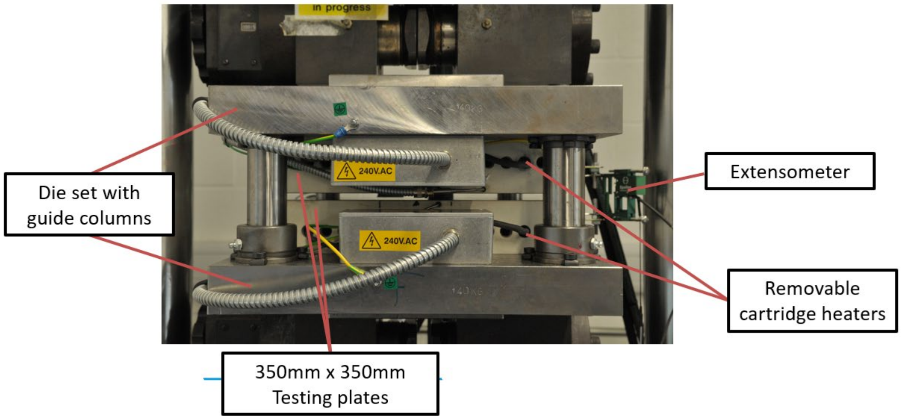
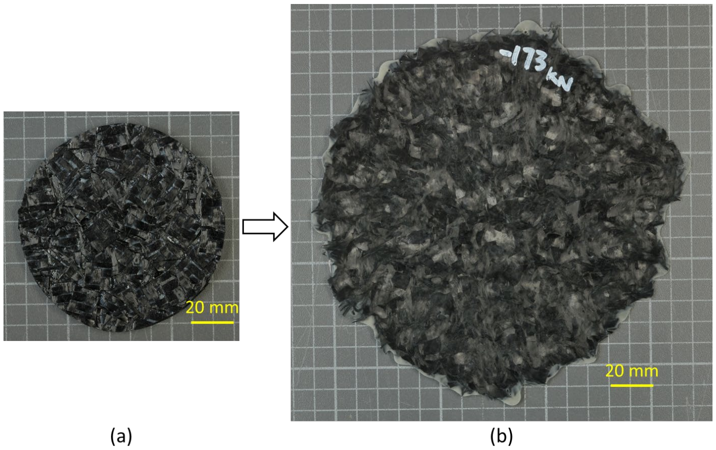


Fig. 2: The squeeze flow rig employed for flow characterisation [3]



(a) (b)

Fig. 3: An 12.5 mm, 3000 gsm chip-SMC specimen (a) before and (b) after the squeeze flow test

Hybrid Architecture Composites Characterisation. Hybrid architecture specimens were also characterised using the squeeze flow rig, where 17.5mm square chip-SMC with 3000gsm areal mass were chosen for all specimens. For these hybrid architecture tests , chip-SMC specimens of 100mm diameter were centrally located on top of two plies of square prepreg with 200mm edge length (see **Fig. 4**). Two different configurations were studied, as shown in **Fig. 4**: one with continuous prepreg in both plies, and the other had four 100mm x 100mm prepreg patches with a series of butt joints in the top ply The smaller prepreg patches were introduced as a third type of architecture which could also be produced using prepreg offcuts, and the characterisation aimed to understand how they affect the potential distortion in the continuous fibre prepreg ply beneath and the flow behaviour of the chip-SMC above.

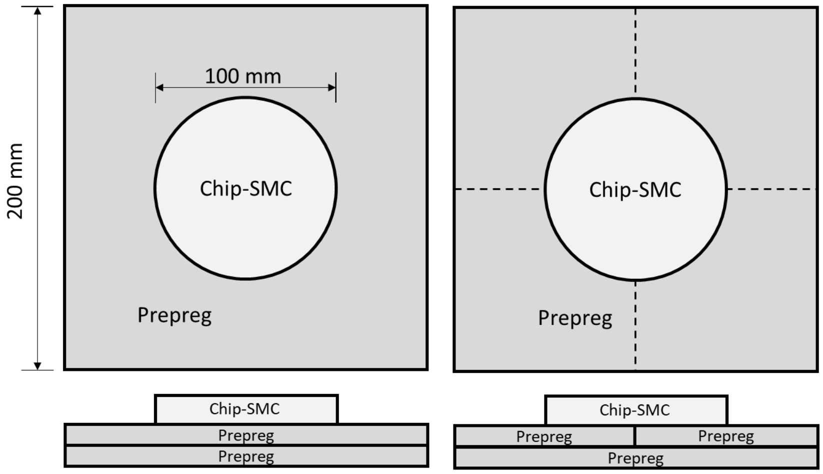


Fig. 4: Specimen configurations for hybrid architecture composites characterisation

Material Characterisation Results

Chip-SMC Characterisation. Fig. 5 presents the area increase after testing for each chip size and nominal areal mass, where the dotted red line indicates the area increase of the baseline SMC specimen. Area increase is a good indicator for the material's ability to flow, which is typically dominated by two deformation mechanisms: compaction and spreading of tows and dispersion of tows. The area increase measures the level of dispersion of tows. However, it should be noted that area increase cannot be directly compared between specimens of different masses, because specimen with larger mass contains more tows, which naturally leads to higher levels of dispersion. According to Figure 5, at the same nominal areal mass, specimens with smaller chips generally experienced larger area increase after testing. The 3000gsm specimens with chip sizes of 12.5mm and 17.5mm both had similar area increase to the baseline SMC (around 200%). However, as the area increase generally decreases with reduced specimen mass, it would be expected that these chip-SMCs would experience lower area increase at an areal mass similar to that of the baseline SMC (1048gsm). This could be explained by the mesoscale fibre architecture: the interlacing tows in chip-SMCs can prohibit the movement of tows, leading to lower level of fibre dispersion. It is also interesting to note that the 17.5mm chip size had 50% more area increase compared to the 25mm chip size, while, the 12.5mm chip size had only 20% more area increase than the 17.5mm chip size. This suggests the possible existence of a threshold for chip sizes below which the material's ability to flow could not be improved anymore. It should be noted that this conclusion is exclusive to square chips manufactured using the specific woven prepreg, other parameters such as chip aspect ratio and the fibre architecture in the prepreg materials can also affect the flow behaviour of chip-SMC, which will be investigated in future work.

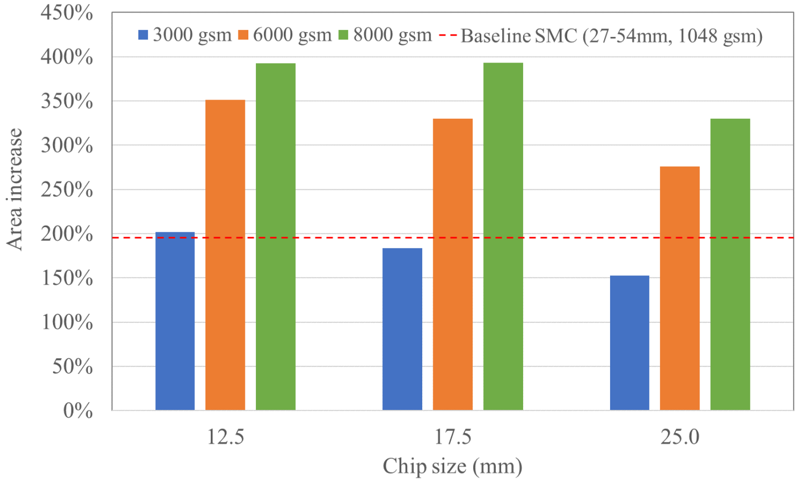


Fig. 5: Comparison of specimen area increase after squeeze flow testing for chip-SMCs of different chip sizes and nominal areal masses. The red dashed line indicates the baseline SMC of 27mm-54mm fibre length and 1048gsm fibre areal mass.

Fig. 6 shows the compressive stress-strain relationship obtained from the squeeze flow test for all chip-SMC specimens. The doted red curve is the stress-strain result for the baseline SMC. Specimens with higher areal mass exhibited lower resistant to compressive pressures during testing, possibly because thicker specimens promote higher level of out-of-plane orientation in chips, resulting in higher level of lofting between chips and leading to more voids. Interestingly, the chip size had less effect on the material's stress-strain response, as curves for specimens with the different chip sizes but same areal mass are grouped together, although specimens with larger chip sizes tend to exhibit up to 35% higher compressive stresses at higher compressive strains. In addition, the curve for the baseline SMC sits closely between the curves for 3000 gsm chip-SMC with 12.5 mm and 17.5 mm chips where the three materials also had similar area increase as discussed above.

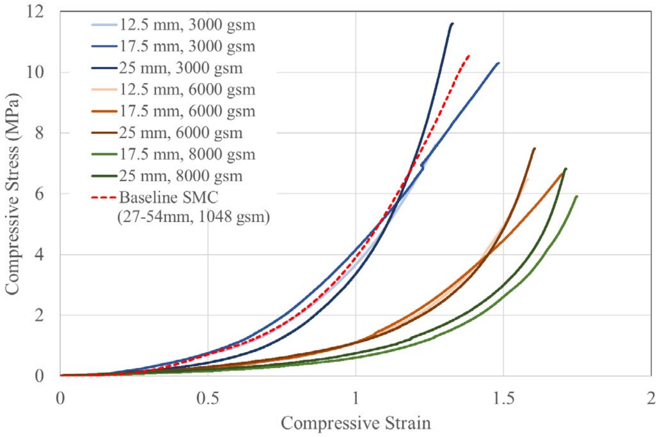
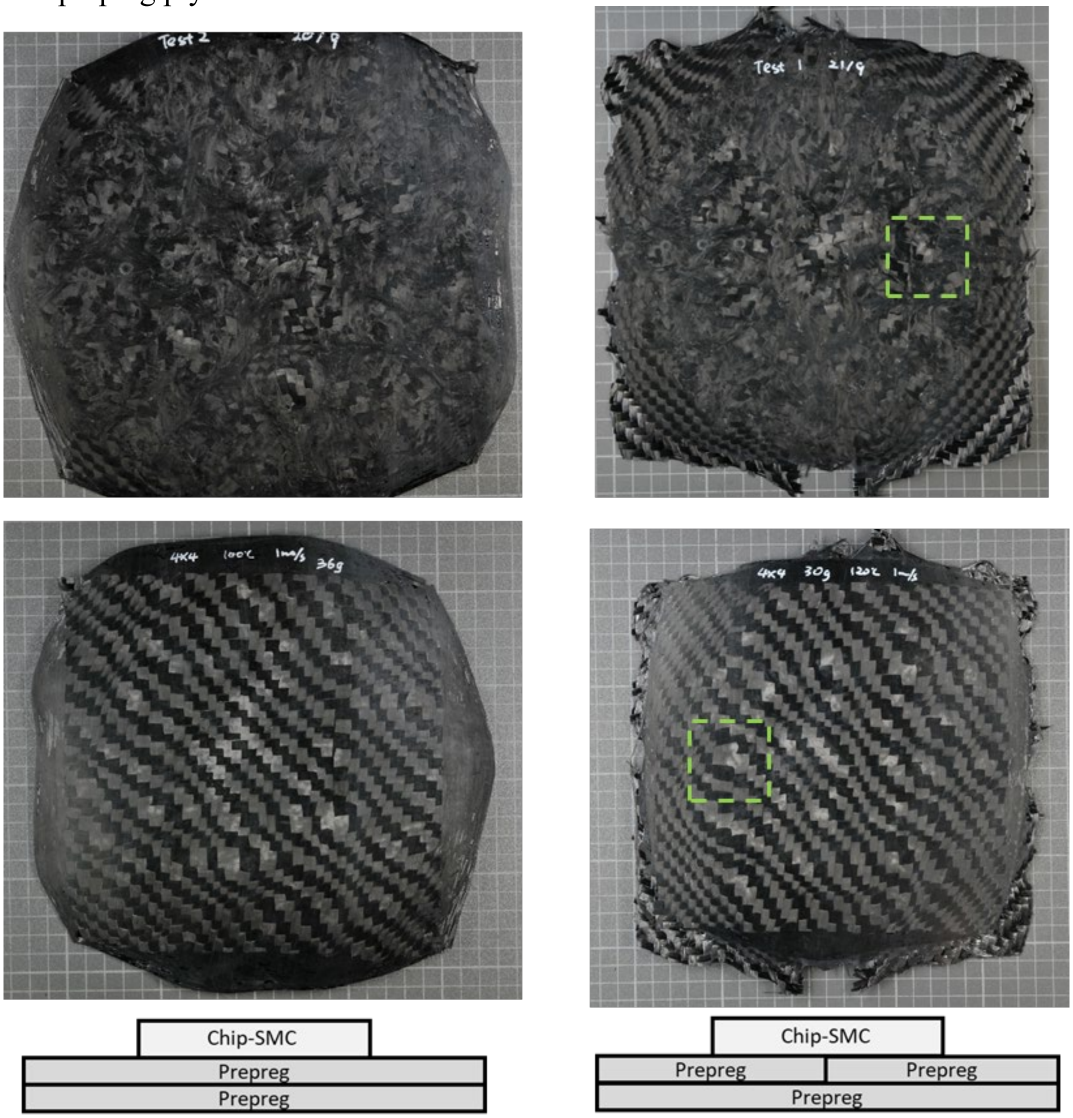
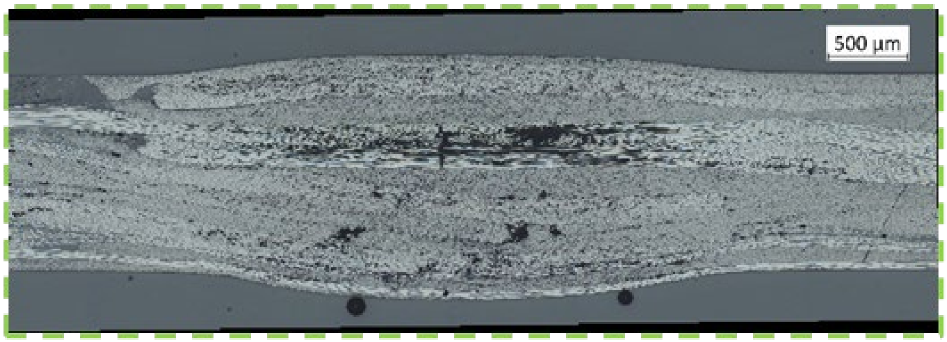


Fig. 6: Comparison of compressive stress-strain response obtained from squeeze flow testing for chip-SMCs of different chip sizes and nominal areal masses. The red dashed line indicates the baseline SMC of 27mm-54mm fibre length and 1048gsm fibre areal mass.

Characterisation of Hybrid Architecture Composites. Fig. 7 (a) presents images of the hybrid architecture specimens after squeeze flow testing, where the top row shows the chip-SMC face and the bottom row shows the continuous prepreg face. Comparison to the chip-SMC face suggests that the prepreg patches restrict the flow of chip-SMC, such that the chip-SMC flow front was further ahead in the specimen without patches compared to the specimen with patches. Areas with compacted tows can be observed ahead of the flow front of chip-SMC in both specimens, although it is less obvious in the specimen without patches as the flow front of the chip-SMC reached the edges of the prepreg plies in most areas. Further research will be performed using larger prepreg plies so that this effect can be better investigated. According to a similar study where conventional SMC was used [3], tow compaction was expected to be accompanied by tow spreading in the area underneath the chip-SMC, which can be seen in the bottom prepreg ply in the prepreg-side images in Fig. 7 (a). It was also reported in [3] that tow spreading in the continuous fibre phase was primarily caused by the compressive stresses arisen from SMC flows, and the effects of friction at the interface were negligible, which was assumed to be the same case in this paper. While the prepreg patches affected the flow of chip-SMC, it had little influence on the level of distortion in the bottom prepreg ply.



(a)



(b)

Fig. 7: (a) Hybrid architecture specimens of two different configurations after squeeze flow test and (b) the cross-section of a highly distorted location (highlighted in (a))

Fig. 7 (b) shows the micrograph of the cross section of an area where extremely large distortion in the bottom prepreg ply was observed, as indicated by the green boxes in Fig. 7 (a). It is interesting to note that tows remain interlaced in the chip-SMC at the same location, alongside an internal crack seen in Fig. 7 (b). A possible explanation is that the fibres were trapped at this location, prohibiting the break-up of the interlaced tows, resulting in higher local fibre volume fraction. The higher local volume fraction led to higher local compressive stresses in the chip-SMC and therefore higher levels of tow spreading in the prepreg underneath. At the same time, there was no longer sufficient resin to bind the fibres together, hence an internal crack was formed once the mould pressure was released.

Compression Moulding of Hybrid Architecture Composites

Compression moulding of hybrid architecture composites were performed to further characterise the manufacturing process in more representative scenarios, as well as evaluate the chip-SMCs' abilities to flow out-of-plane, which could not be assessed using in-plane squeeze flow testing. All chip-SMC used in the compression moulding studies consisted of 25mm square chips. A flat plaque geometry of 200mm x 300mm x 2.5mm was chosen for the compression moulding studies.

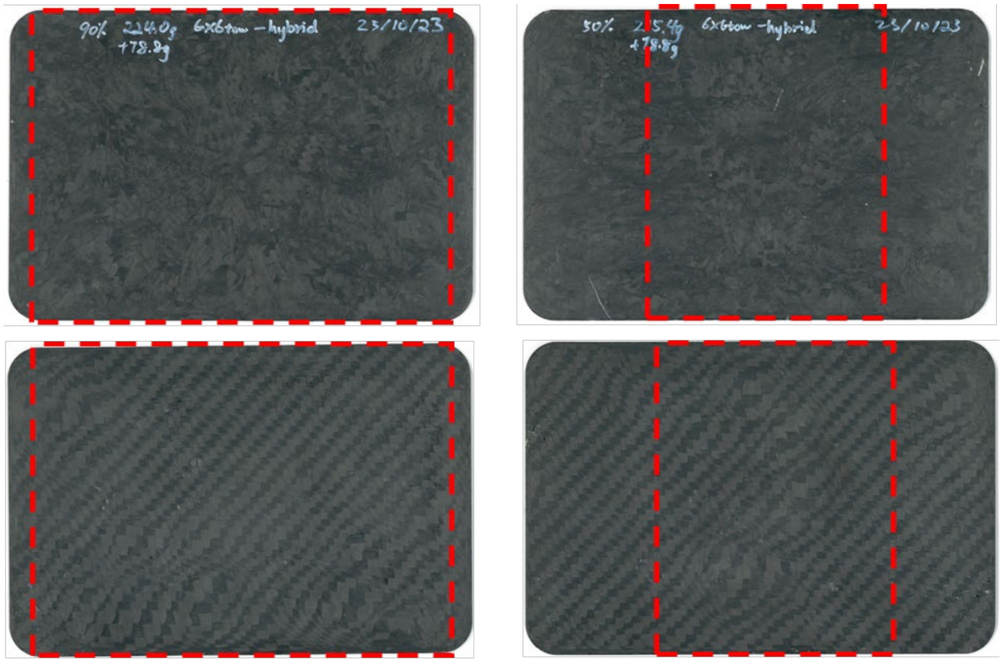
Fig. 8: Prepreg plies and chip-SMC charges in the flat plaque mould cavity on the Langzauner press

Flat plaques were moulded using a Langzauner 150 Tonne multifunctional fast-closing press. Centrally located rectangular initial charges were employed to introduce a 1D flow regime (Fig. 8), using an initial charge coverage of 90% or 50% . For both initial coverage sizes, 2 plies of net-shaped prepreg were used and the prepreg plies were positioned directly on the mould surface as shown in Fig. 8. It should be noted that unlike the material characterisation studies where the chip-

SMC samples were cut from a larger sheet, the initial charges for compression mouldings were manufactured net-shaped. In addition, no compaction was performed to the initial charges. Consequently, the initial charge had high variation in thickness distributions, and strong edge effects such that the areal mass in the centre was significantly higher than around the edges (see Fig. 8).

Fig. 9 presents the images of the moulded flat plaques, where the top row shows the chip-SMC side and the bottom row shows the prepreg side. For both 90% and 50% initial coverages, the chip-SMC achieved complete filling, since no prepreg was visible from the SMC side of the images. Unbroken chips and interlaced tows can be observed in both plaques, particularly around the centre of each plaque where the initial charge was originally located. This indicates a difference in flow mechanism for the chip-SMC compared to conventional SMC. In conventional SMC, the fibre architecture characteristics do not have noticeable variations along the flow direction [4], meaning the material experiences uniform local deformations. The chip-SMC on the other hand experiences higher flow towards the edges of the initial charge, and lower flow in the centre of the initial charge.

It is also interesting to observe from the prepreg-side in Fig. 9 that the high levels of tow spreading in continuous fibres were mainly observed in the areas directly underneath the initial charges, as indicated by the red boxes. This is because the higher thicknesses and areal masses towards the centre of the initial charge led to higher resistance to compression, resulting in higher compressive stresses applied to the prepreg.



90% coverage

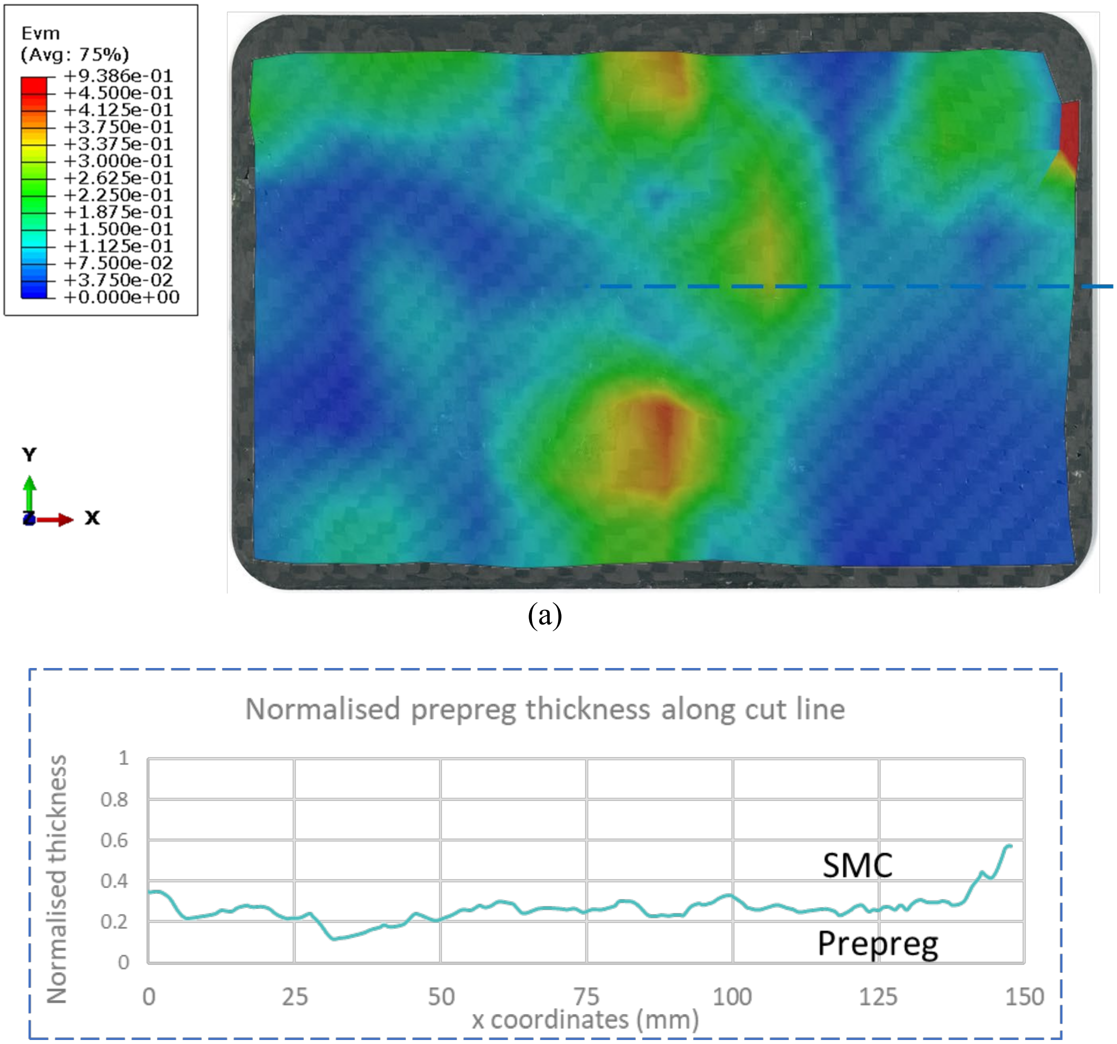
50% coverage

Fig. 9: Hybrid architecture flat plaques moulded from two different initial charge coverages (highlighted)

Fig. 10 (a) presents the in-plane strain distribution in the prepreg ply for the 50% initial coverage plaque, obtained using the grid strain analysis (GSA) method proposed in [5]. The colour contour shows the equivalent strain, calculated as:

$$\varepsilon_{eq} = (1/\sqrt{2})[(\varepsilon_x - \varepsilon_y)^2 + (3/2)\gamma_{xy}^2]^{1/2}. \quad (1)$$

where ε_x, ε_y and γ_{xy} are in-plane strain components. Higher strains were mainly observed around the centre of the initial charge, which was in agreement with the higher levels of the tow spreading observed in Fig. 9. Fig. 10 (b) presents the thickness distributions in the chip-SMC layer and prepreg along the cross section indicated by the blue line in Fig. 10 (a), where the thickness values were manually measured using micrographs of the cross-section. The 0 x-coordinate in Fig. 10 (b) indicates the geometric centre of the plaque. In general, higher SMC/prepreg thickness ratios were observed in areas with higher levels of tow spreading, therefore the SMC/prepreg thickness ratios show a decreasing trend from the centre (x=0) to the edge (x=150mm) of the plaque.



(a)

(b)

Fig. 10: (a) Area change of grid elements calculated with grid strain analysis method and (b) thickness distribution of two fibre architectures (SMC and prepreg) along the dashed line indicated in (a)

Conclusions

Characterisation of chip-SMC materials have shown that smaller chip sizes experience greater levels of area increase during squeeze flow.However, chip-SMCs general experience less area increase compared to the baseline SMC as the interlaced tows prohibit fibre dispersion. Analysis of the compressive stress-strain relationship has suggested that the material's resistance to compression stresses decreases with increasing areal mass, but chip size has little influence on the compressive stress-strain response.

Characterisation of chip-SMC/prepreg hybrid architecture have shown that the flow of chip-SMC introduces tow spreading in the area underneath the SMC, and tow compaction in the area ahead of the flow front of the SMC. An additional study has been conducted to investigate the effects of introducing a third fibre architecture consisting of prepreg patches between the chip-SMC and the continuous prepreg ply. While no significant effects are observed on the level of distortions in the continuous prepreg ply, the prepreg patches experience large distortion and the flow of SMC is restricted, suggesting a reduced overall formability and mechanical properties compared to the hybridisation of chip-SMC and continuous prepreg only. Higher levels of tow spreading and internal cracks tend to appear in areas where interlaced tows remain in the chip-SMC.

Finally, compression mouldings of hybrid architecture composites have been performed using a flat plaque geometry. Variations in the fibre architecture in the chip-SMC and the level of tow spreading in the continuous prepreg have been observed using different initial coverage sizes. These are due to the higher thickness and areal mass variations in the larger initial charges, compared to the smaller specimens used in the material characterisation studies.

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