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# **Diverted from Landfill: Manufacture and characterisation of composites from waste plastic packaging and waste glass fibres**

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## **Abstract**

This work explores the use of low-value packaging film-based waste mixed plastics (wMP) and waste glass fibres (wGF) to produce value-added composites. The study involves producing thermoplastic prepregs with wMP and wGF, manufacturing laminates via compression moulding, optimising wGF content, analysing the interface and assessing the performance of the laminates through mechanical testing. The results indicate that adding 18 - 26 vol% wGF to unreinforced wMP leads to significant improvements in tensile strength (over 300%), tensile modulus (500%), flexural modulus (over 700%), compressive strength (~350%), and compressive modulus (~700%) compared to unreinforced wMP. The significance of having 2D dispersion of short fibres with partial orientation compared to the more conventionally used 3D dispersion of short fibres is discussed. The research provides valuable scientific insights into the application of mixed waste materials in composites, aiding the creation of a more circular economy for plastic waste and leading to new composite products.

Keywords: mixed waste plastics; recycled plastics; waste glass fibres; compression moulding, waste-based composites; polyethylene blends

## 1. Introduction

Plastic packaging films play an important role in protecting goods and reducing food waste, but their recycling system is not sufficiently developed. The plastic packaging consisting of high-density polyethylene (HDPE), polypropylene (PP), and polyethylene terephthalate (PET) are commonly recycled. A large proportion of soft plastic packaging wastes, that consist primarily of low-density polyethylene (LDPE), are not generally recycled due to their flexibility, low mechanical performance and low cost [1]. These thin plastic films originating from packaging are typically single-use and have a short life span, and they often end up in landfill once their initial purpose is fulfilled, creating significant environmental challenges. Within the UK, it is estimated that five million tonnes of plastics are used every year [2], with 67% of this used for packaging [3]. Thin LDPE films and bags are not collected with household recycling in the UK, but their inclusion could significantly increase the recycling rates. If both thin plastic films and common household recycling were systematically collected and processed as a unified stream, that could eliminate the necessity for segregating plastics in the recycling process, leading to cost savings.

It is important, however, to understand the effect of polyolefin blends on the mechanical properties compared to those of the individual polymers. Studies investigating polyethylene (PE) blends have mostly considered the effects on virgin materials. There have been some studies on blending recycled PEs/polyolefins in different proportions. Yousif et al. [4] studied different blends of recycled LDPE (rLDPE), rHDPE, and rPP. Blending the three with different proportions was shown to increase the tensile stress up to 14-25% compared to the individual polymers alone, confirming the potential increase in the quality of recycled plastics through blending. Similarly, Cecon et al. [5] investigated the effects of adding different amounts of post-consumer recycled polyethylene (PCRPE) with virgin PEs of different densities (vLDPE), virgin linear LDPE (vLLDPE), virgin medium density PE (vMDPE), and vHDPE. Both vLDPE and vLLDPE blends displayed increases of up to 75% in the tensile modulus and 56% in the tensile yield strength compared to the individual polymers.

Waste mixed plastics (wMP) originating from soft plastic packaging films consist primarily of various grades of PE, including both LDPE and HDPE. From the aforementioned studies, it is evident that such wMP has the potential to offer mechanical properties superior to LDPE alone; this is due to the presence of higher-strength polyolefins in the mix [5, 6]. The mechanical properties of polyolefin blends can be increased further through reinforcement with glass fibres (GF). There have been many studies investigating the effect of adding GF reinforcement to mixed polyolefin blends [7-11], and to individually recycled polyolefin matrix composites with various reinforcements [12-18] such as wood [14, 17-20], fly ash [15, 21], and glass [13, 14]. To the best knowledge of the authors, there has not been any study focusing on the effect of the addition of GF to a polyolefin blend containing only waste plastics.

In Alqaflah's research [7] GF-reinforced composites were manufactured using a blend of PET and LLDPE as the matrix. This study revealed that the incorporation of a blend matrix results in a composite material that exploits the favourable properties of both polymers. The incorporation of LLDPE, known for its lower tensile strength relative to PET, led to a reduction in the tensile strength of the blend compared to neat PET, and concurrently increased impact resistance and toughness. The increase was more pronounced at higher GF loadings, up to 30

wt%. Additionally, the utilisation of a blend matrix did not adversely impact the interface between the matrix and the fibre. It is worth noting that the benefits of using blends are mainly investigated with higher-performing plastics like HDPE, LLDPE, and PET. Very few studies focus on low-performing plastics like LDPE or LDPE blends. In one such study, Bajracharya [9] focused on lower-performing plastic blends, including waste LDPE, HDPE and PP of unknown proportions, reinforced with virgin short GF (vGF). This investigation showed that the addition of vGF resulted in significant improvements in strength and elastic modulus, with the best improvement seen in flexural properties. The paper mentions a few limitations, including the challenges regarding the random orientation, and length of the vGF in predicting tensile modulus and strength properties, and the potential formation of voids due to entrapped air and moisture within the compounded pelletised materials. Additionally, in both studies [7] and [9], the vGF loading up to only 30 wt% was investigated; hence the benefits and drawbacks of higher GF contents were not explored.

As mentioned previously, composites containing polyolefins and short GF are generally manufactured through melt mixing via extrusion. This ensures a homogenous composite; however, extrusion further shortens the length of the fibres, which can affect the mechanical properties of the resulting composite. Ayadi [12] noted that after one extrusion of short vGF with vHDPE, the viscosity of the composite increased, the fibre length decreased, and the decrease in fibre length was more pronounced as the GF proportion increased in the composite. This fibre length reduction was also reported in Bajrachayra's investigation [9].

The reduction in fibre length can be avoided if the composite is produced as a laminate where the short GF are in non-woven mat form and do not undergo high shear during melt impregnation. It is well known that the manufacture of composites using compression moulding results in lower shearing stress and lower breakage of GF [22, 23], compared to extrusion. Additionally in extrusion, the increasing viscosity of the melt with increasing GF loading contributes to poor fibre dispersion [24], collectively leading to a poor-quality composite. This might lead to slightly lower mechanical properties in the extruded composites (with 3-D fibre dispersion) compared to that in an equivalent laminate (with 2-D fibre dispersion) with a similar loading of short GF.

In the context of existing literature, it is notable that a substantial portion of prior research within this domain focuses on extruded or injection moulded short GF-reinforced composites [7-9, 12-14, 25-29]. Various justifications for utilising extrusion in the manufacturing of short GF-reinforced composites were presented in the published papers citing its economic efficiency and continuous nature of processing [29]. Extrusion also facilitates the addition of additives or compatibilisers [30]. These studies predominantly explored composites where the orientation of fibres is inherently random due to the extrusion process. In contrast, the integration of GF plies within a laminate configuration results in a two-dimensional (2-D) fibre arrangement as opposed to the three-dimensional (3-D) fibre distribution commonly observed in extruded composites. Also, the shift in fibre distribution within the composite, 2-D in comparison to the more conventional 3-D, highlights the novel perspective presented in this study, particularly in relation to short fibre reinforced thermoplastic composites. Moreover, the papers discussed above focused on investigating GF loadings of up to 30 wt%, with only a limited number of studies extending their investigations up to 40 wt%. In the current study, GF loadings of up to 44 wt% were investigated.

The majority of soft plastic film waste ends up in landfill once it has served its initial purpose in the UK. About 150000 tons of GF end up in landfill in the EU each year [31], and approximately 67% of GF is sent to landfill in the UK each year [32]. The addition of waste GF to waste plastics can convert these disposable materials into value-added composites suitable for a range of applications, such as construction [9] and consumer products [33]. Bajracharya [9] demonstrated that vGF-reinforced wMPs have strength properties comparable to timber, indicating their suitability for construction applications. Hence, economically viable technologies are needed to convert these wastes into product forms. This study addresses this gap and proposes a technology that can divert two wastes from landfill to market in suitable product forms. The novelty of the work lies in the combination of two wastes, resulting in a consistent quality starting material (thermoplastic prepreg) for composite manufacturing. This approach ensures a uniform 2-D dispersion of wGF in composites and minimises variability arising from different wMP batches with varied compositions.

In this study, wGF was coated with wMP via extrusion coating to produce consistent quality thermoplastic prepregs. The produced wMP/wGF prepregs were used to manufacture laminates via compression moulding. The viscosity of the wMP was measured using rheometry to decide the compression moulding temperature. The wGF content was varied in the laminates, and compression moulding parameters were optimised. Tensile, flexural, and compressive tests were conducted to evaluate the mechanical properties of the laminates. Additionally, scanning electron microscopic (SEM) images were captured to assess the quality of consolidation within the laminates and the wGF/wMP interface.

## **2. Experimental:**

### **Materials**

#### *Waste mixed plastics*

The primary materials of focus in this study are waste mixed plastics (wMP) and waste glass nonwoven mats (wGF). The industrial collaborator PALTECH (Polymer Alloy Technology) [33] takes the plastic wastes originating from food packaging from Tesco Ireland supermarkets [34]. These mixed plastic packaging wastes undergo one stage of sorting, which is a float-sink separation. The wMPs that float on water are collected by PALTECH. Therefore, the wMP predominantly consists of polyolefins with densities lower than that of water. The wMP is washed, shredded, and supplied for this research by PALTECH in the form shown in Figure 1a. Differential scanning calorimetry (DSC) studies carried out in our previous work [6] revealed that the polyolefins found in the mixture predominantly consist of various grades of PEs.

#### *Waste glass fibre*

Johns Manville, one of the industrial collaborators, supplied the wGF roll, which was one of many out-of-specification rolls that are routinely sent to landfill. The wGF roll consists of a uniform non-woven GF mat with an organic binder (urea formaldehyde) present on the GF surface, having a weight of approximately 23 wt% with respect to the weight of GF. The wGF mat has an areal weight of 100 gsm (with binder) (DH 100) [35], a nominal fibre length and diameter of 18 mm and 13  $\mu$ m respectively, and the roll had a width of 350 mm. The wGF

nonwoven mat used in this study was not 100% random but had a different fibre directionality along the length of the mat ( $0^\circ$ , which is the machine direction) and across the length ( $90^\circ$ , which is the cross machine direction), indicated in Figure 1b. This directionality is due to the manufacturing method of the wGF rolls. In the wGF used here, there are more fibres aligned towards the machine direction (MD) than in the cross machine direction (CMD) and the MD:CMD tenacity ratio is 1.8 [35].

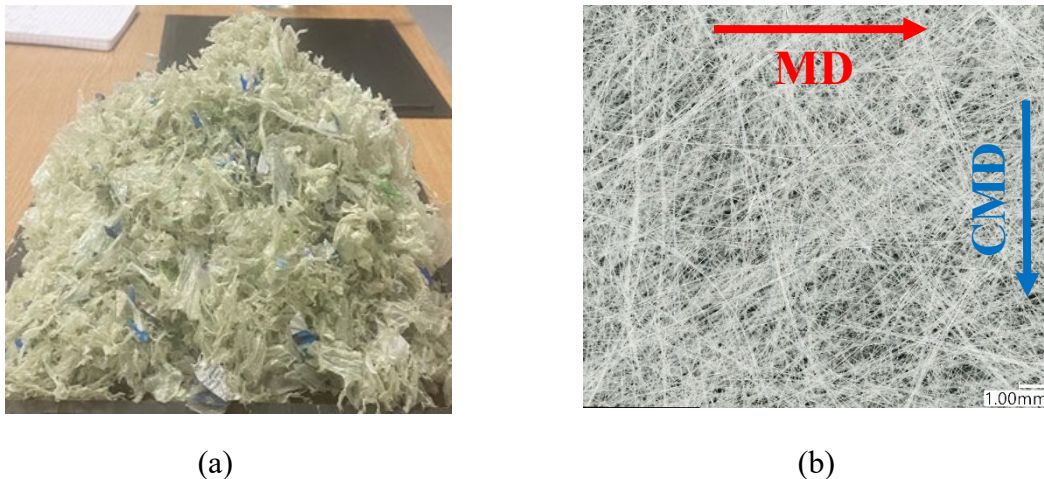


Figure 1: (a) Waste mixed plastics (wMP) in shredded form, and (b) waste glass fibre (wGF) nonwoven mat.

## Manufacturing

The work originally began with shredded wMP and shredded wGF, with the composites manufactured solely by compression moulding. The resulting composites, however, showed a non-uniform distribution of wGF. A laminate manufacturing route was then adopted with 2-D dispersion of short GF using wGF nonwoven mat rather than shredded wGF. The wGF was coated with wMP via extrusion coating to produce wMP/wGF prepregs. The adoption of this technique successfully produced uniform quality wMP/wGF prepregs for laminate manufacturing. As the wGF used in this study has a low areal weight (100 gsm), that facilitated good impregnation of wMP within the wGF nonwoven mat. The extrusion coating was carried out using a Killion extruder with a 600 mm wide sheet extrusion die. The processing temperature ranged from  $180^\circ\text{C}$  -  $200^\circ\text{C}$  and the screw speed was set to 30 revolutions per minute. The thickness of the wMP layer on the wGF varied between 0.95 – 1.35 mm. The coating of wGF with wMP is a continuous process, as shown in Figure 2a below, and can be easily translated to an industrial scale.

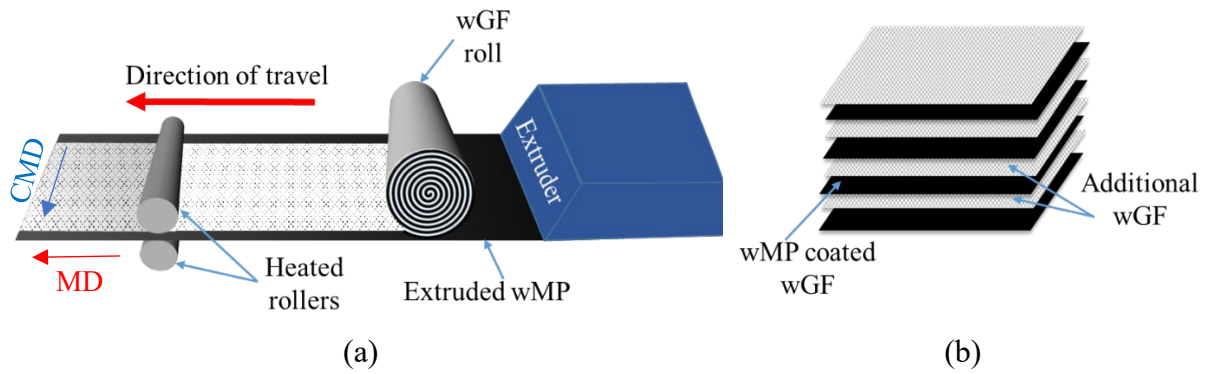


Figure 2: (a) Continuous extrusion coating process to create a waste mixed plastic (wMP)/waste glass fibre (wGF) roll and (b) waste mixed plastic-coated waste glass fibre laminate stacking with additional dry waste glass fibre plies.

The wGF/wMP prepreg produced by extrusion coating is shown in Figure 3. The prepreg was cut to the required laminate dimensions, layers were stacked one above another and compression moulded to produce wMP/wGF laminates. Additional plies of wGF, in uncoated and as-received form, were added to increase the GF content of the laminate as required, shown in Figure 2b.



Figure 3: Manufactured roll (thermoplastic prepreg) of waste mixed plastic (wMP) coated waste glass fibre (wGF).

Laminates were manufactured by compression moulding with a Pinette Emidecau Industrie (PEI) lab 450 hydraulic press, using a two-piece metal mould with inner cavity dimensions of  $350 \times 300 \times 2$  mm. The mould is shown in Figure S1. The dry wGF plies and wMP-coated wGF plies were stacked to fulfil various fibre volume fractions (FVF), calculated based on the weights of each component. The stacking arrangement is depicted in Figure S2. The impregnation of wMP through the wGF plies during the pressing stage, however, became progressively more challenging with an increasing number of dry wGF plies, thereby leading to a variation in the consolidation pressure requirements, as shown in Tables 1 and 2.

The processing cycle used in the hydraulic press was as follows:

1. Heating from 20°C to 200°C at a rate of 10°C /min.
2. Holding at 200°C for 10 minutes at 5 – 25 bar (Table 1 and 2).
3. Cooling from 200°C to 20°C at 10°C /min.

The processing temperature of 200°C was decided based on the thermogravimetric analysis (TGA) carried out in our previous work [6], and from the rheometry carried out in this study, described in section 3.1. Eleven laminates were manufactured and test samples were extracted from them in both MD and CMD for various mechanical testing. The details of all the laminates are given in Tables 1 and 2.

## Test methods

### Density

The density of each laminate was measured using an OHAUS density determination kit and following ASTM D792. At least six samples were measured from each set. The following equation was used to determine the density ( $d$ ) of each sample, and an average was taken for each laminate.

$$d_{sample} = \frac{\text{mass in air (g)}}{\text{mass in distilled water (g)}} \times d_{distilled\ water} \left( \frac{g}{cm^3} \right) \quad (1)$$

### Void content

Before measuring the void content, the content of organic binder on the surface of the wGF was measured, by holding samples of dry wGF at 550 °C for 5 hours in a Nabertherm furnace. The dry wGF samples were weighed before and after burning to measure the weight of organic binder present on the wGF surface prior to burn-off.

To measure the void content, samples from each laminate were held in a Nabertherm furnace at 550 °C for 5 hours to remove the matrix completely. At least six samples were measured from each set. The weight of the organic binder was taken into consideration when calculating the FVF of the samples. The fibre volume fraction (FVF) of each sample was calculated following the ASTM standard D3171, using the equation below.

$$FVF = \frac{V_{GF}}{V_{GF} + V_{wMP} + V_{binder}} \times 100 \quad (2)$$

where

$$V_{GF} = \frac{M_{GF}}{\rho_{GF}} \quad (3) \quad V_{wMP} = \frac{M_{wMP}}{\rho_{wMP}^*} \quad (4) \quad V_{binder} = \frac{M_{GF} \times 0.23}{\rho_{binder}} \quad (5)$$

\*The density of the unreinforced wMP was measured to be 0.94 g/cm<sup>3</sup> as reported in our previous paper [6].



### ***Rheology***

Rheological experiments were carried out on a TA Instruments AR-G2 controlled stress rheometer. Shreds of wMP were compacted into 2 mm thick plaques from which 25 mm discs were die-cut using a Dr Collin platen press. Frequency sweeps were carried out from 0.01 to 100 Hz on wMP at temperatures of 160°C, 180°C, 200°C and 220°C using a controlled strain of 0.5%. A Cox Merz model was applied to the frequency sweep data to predict the shear viscosity data.

### ***Tensile testing***

Tensile testing was used to determine the tensile strength and modulus of each type of laminate. Six samples were tested from each laminate on an Instron test frame fitted with a 10 kN load cell in accordance with ASTM D3039. The test samples were cut from each laminate using a guillotine. Each sample was tested at a speed of 2 mm/min until failure, and the values for the load and extension were recorded using the Bluehill® testing software.

### ***Flexural testing***

Flexural testing was carried out following ASTM D790, using six samples from each laminate. The test speed used was 5.12 mm/min on an Instron 3369 test frame fitted with a 10 kN load cell, and the span-to-thickness ratio was 16:1. The load and deflection values were recorded using the Bluehill® testing software. Only the flexural modulus was measured as the samples did not break during testing, which is required to measure the flexural strength.

### ***Compression testing***

Compression testing was carried out following ASTM D6641 using six samples from each set. The test speed was 1.28 mm/min on an Instron 2269 test frame fitted with a 10 kN load cell, and the strain was measured using a video extensometer. Values for the load were recorded using the Bluehill® testing software.

### ***Microscopy***

A scanning electron microscope (JEOL JSM series SEM) was used to assess the quality of consolidation and to examine the voids present in each laminate. The cross sections of the samples were cast in epoxy, polished, and were prepared by gold sputtering alongside the fracture surfaces, to increase the conductivity. The voltage used was 15 kV.

A Keyence VHX-6000 Series 3D imaging microscope was used to capture high-resolution images of the wGF mat to observe the orientation of the fibres.

## **3. Results and Discussion**

### **3.1 Rheometry**

Figure 4 shows the viscosity of wMP as a function of shear rate at four different temperatures: 160°C, 180°C, 200°C, and 220°C.

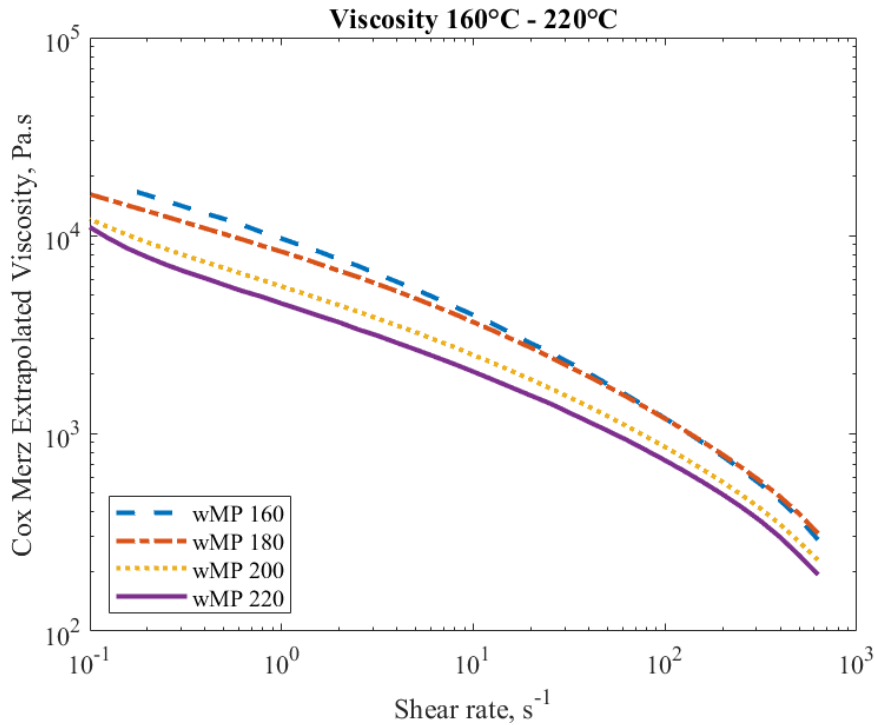


Figure 4: Graph showing the Cox Merz Extrapolated Viscosity against the shear rate for waste mixed plastic (wMP) at 160°C, 180°C, 200°C, and 220°C.

The typical shear rate of thermoplastics during compression moulding is approximately 1 – 10 s<sup>-1</sup> [36]. The wMP samples exhibited a drop in viscosity with the increase in shear rate, as expected with thermoplastic polymers. The viscosity profiles at 160°C and 180°C were very similar, as seen in Figure 4. There was an 18% drop in viscosity with further rise in temperature, and the viscosity profiles at 200°C and 220°C were quite similar. Based on these observations, the laminate manufacturing temperature was decided to be 200°C to ensure good impregnation with lowered viscosity whilst avoiding any detrimental thermal degradation [6].

### 3.2 Density and Void content

Eleven laminates were produced with different volume fractions of wMP and wGF, six of which with samples cut in the MD and five with samples cut in the CMD. Descriptions of the laminates are provided in the table below.

Table 1: Summary of measurements of manufactured laminates with samples cut in the machine direction.

Laminate ID (MD)	wGF content (vol%)	wMP content (vol%)	Binder content (vol%)	Laminate Density (g/cm <sup>3</sup> )	Void content (vol%)	Moulding Pressure (bar)	Thickness (mm)
L6	5.5	73.1	2.1	0.86	19.3	5	2.91
L12	11.5	78.5	4.5	1.10	5.49	5	3.69
L18	18.3	70.5	7.2	1.24	3.97	15	2.89
L23	23.5	54.8	9.2	1.25	12.57	25	3.18

L25	25.0	54.8	9.8	1.30	10.50	25	2.67
L26	26.1	50.5	10.2	1.29	15.04	25	3.27

\* The name of each laminate corresponds to its wGF content, e.g. L6 = 6 vol% wGF.

Table 2: Summary of measurements of manufactured laminates with samples cut in the cross machine direction.

Laminate ID (CMD)	wGF content (vol%)	wMP content (vol%)	Binder content (vol%)	Laminate Density (g/cm <sup>3</sup> )	Void content (vol%)	Moulding Pressure (bar)	Thickness (mm)
L6	5.5	81.6	2.2	0.94	10.73	5	2.81
L12	11.8	79.2	4.6	1.11	4.40	5	3.74
L18	17.8	69.3	6.9	1.21	5.97	15	2.89
L24	24.2	53.5	9.5	1.26	12.78	25	2.95
L25	24.9	55.1	9.7	1.30	10.33	25	2.64

### 3.3 Tensile testing

The measured tensile strengths of the laminates are shown in Figures 5a and 5b.

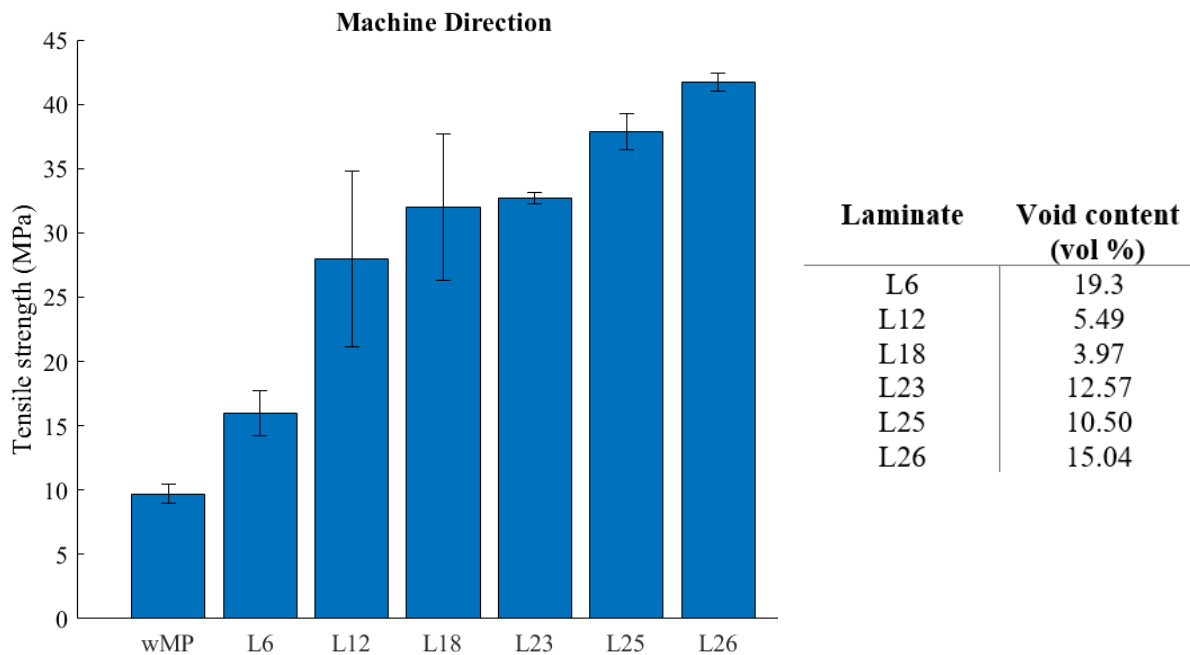


Figure 5a: Comparison of tensile strength between unreinforced waste mixed plastics (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the machine direction (MD).

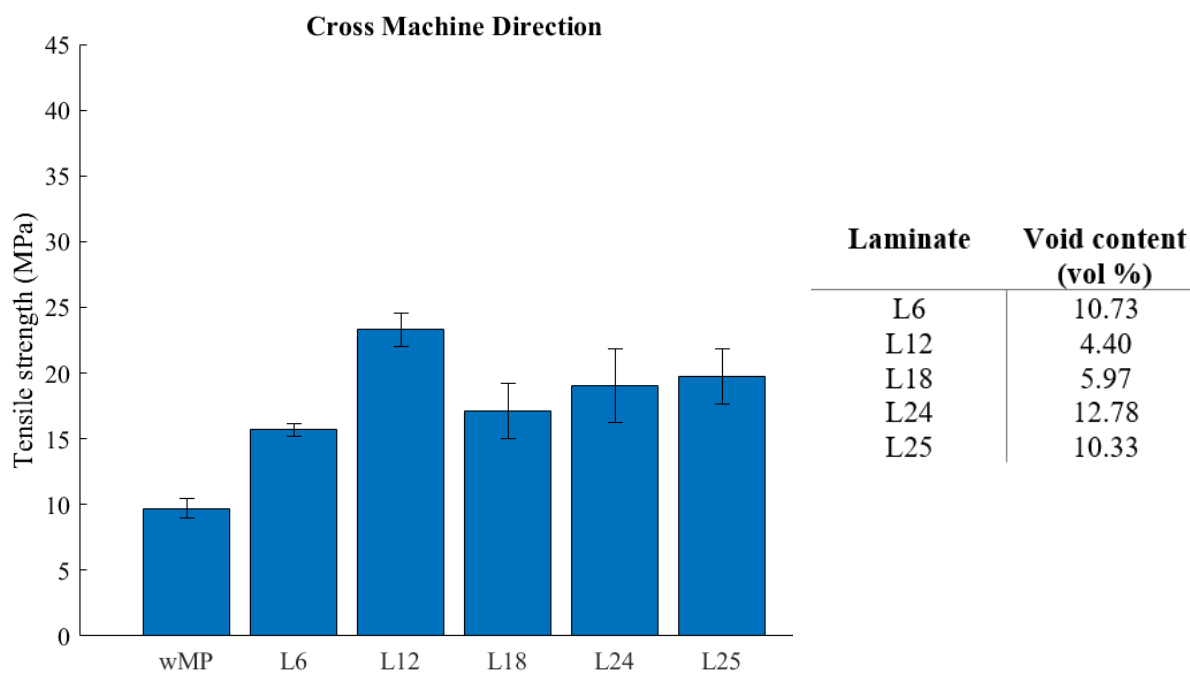


Figure 5b: Comparison of tensile strength between unreinforced waste mixed plastics (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the cross machine direction (CMD).

The highest tensile strength was observed in the L26 (MD) wMP/wGF laminate, with a value of  $41.7 \pm 0.4$  MPa. The addition of 26 vol% wGF to unreinforced wMP increases the tensile strength from 9.7 – 41.7 MPa, an increase of around 330%.

The presence of voids in the laminates has a prominent effect on the tensile strength as seen clearly in the case of L18 (low void content) and L23 (high void content), as shown in Figure 5a. L18 and L23 have similar tensile strengths despite the 5 vol% increase in wGF content; this can be attributed to the higher void content of 12.6 vol% in L23, compared to only 4 vol% in L18. The voids act as stress concentrators and weak points leading to premature failure. The samples cut in the CMD exhibited lower tensile strengths than their equivalent MD samples (Figure 5b). This is due to the lower number of fibres orientated along the test axis during tensile testing in CMD samples, further discussed in section 3.3.1.

Figure 6 compares the tensile strength values of wMP/wGF laminates in this study to GF-reinforced composites with different polyolefin matrices from literature [6, 7, 9, 13, 25, 30]. All the literature values correspond to extruded composites. The wGF content of the laminates in this study has been converted to weight% for ease of comparison to the literature values.

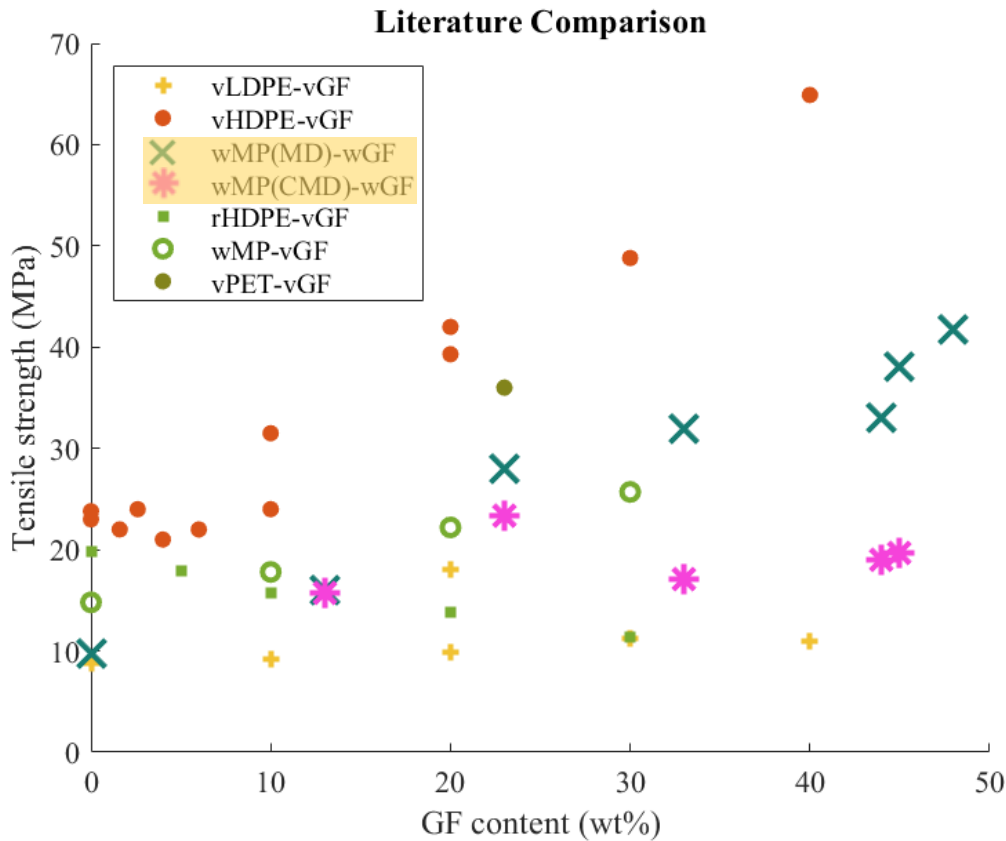


Figure 6: Graph showing the tensile strength of the laminates as a function of waste glass fibre (wGF) content (wt%) compared to the extruded composites with different polyolefin matrices from the literature [6, 7, 9, 13, 25, 30]. The machine direction (MD) and cross machine direction (CMD) results from this study are represented by “×” and “\*” respectively, and highlighted in the legend.

Notably, there are limited studies on extruded composites, i.e. 3D dispersion of fibres, with GF contents exceeding 30 wt%. Increasing the GF content is likely to be more feasible in laminate form, as demonstrated in this current investigation. Figure 6 clearly shows that the MD wGF/wMP results are higher than the vLDPE composite literature values [25, 30], but less than the vHDPE composite values [13, 25]. This is significant as it highlights the benefits of utilising PE-based wMPs over LDPE alone with regard to the tensile properties.

Figures 7a and b show the tensile modulus of the laminates in the MD and CMD, respectively.

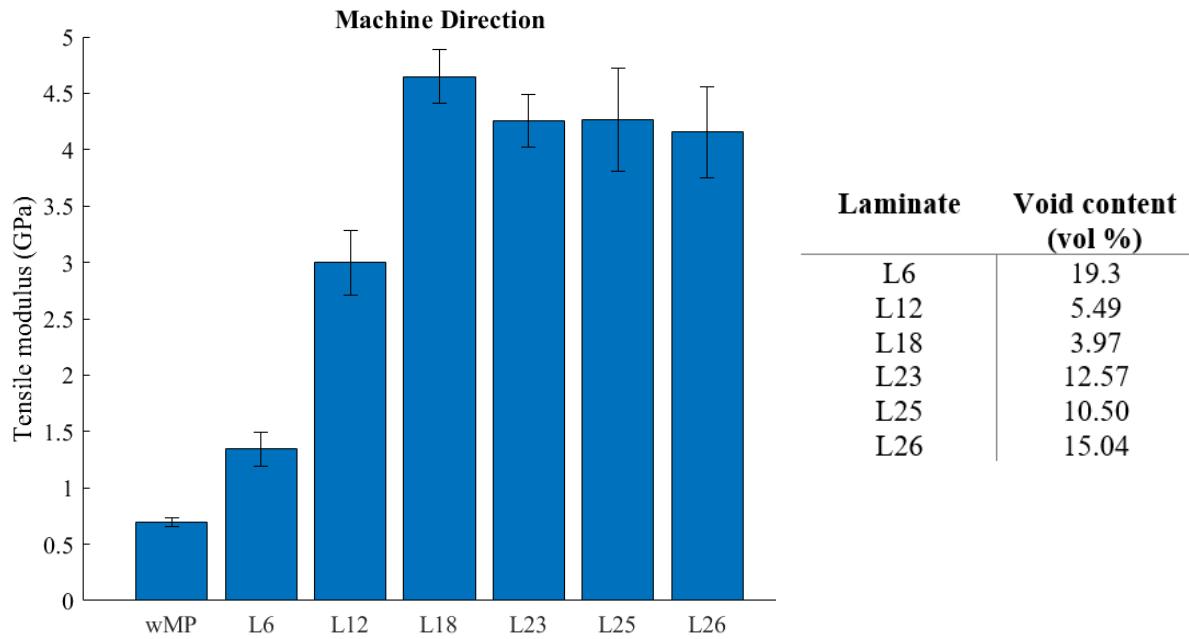


Figure 7a: Comparison of tensile modulus between unreinforced waste mixed plastic (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the machine direction (MD).

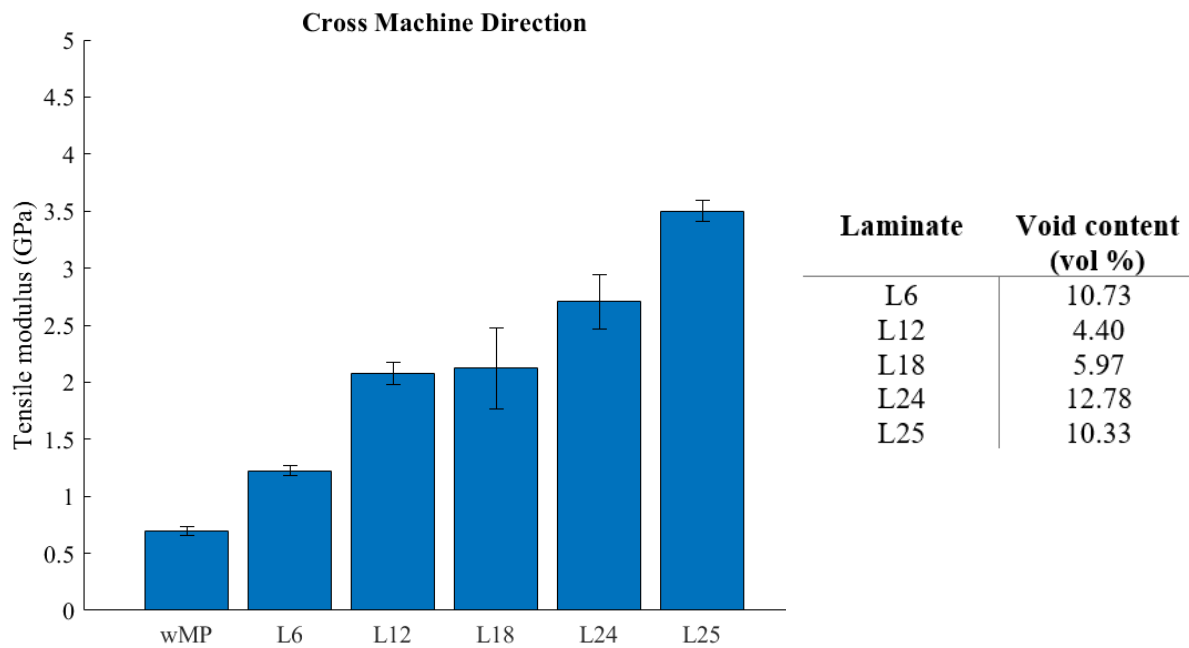


Figure 7b: Comparison of tensile modulus between unreinforced waste mixed plastics (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the cross machine direction (CMD).

The highest tensile modulus was observed with L18 (MD), with a value of  $4.6 \pm 0.2$  GPa (Figure 7a). The tensile modulus values were lower in the wMP/wGF laminates cut in CMD (Figure 7b). This observation verified the fact that there were fewer fibres in CMD that could take up the load during tensile testing. The tensile modulus of L18 (MD) was higher than that

of L23 (MD), L25 (MD) and L26 (MD) (Figure 7a). This anomaly can be explained by the fact that L18 had a much lower void content (4 vol%) compared to L23, L25 and L26 (10-15 vol% voids). This indicates that void content plays a prominent role in controlling the tensile modulus of short fibre composites, unlike that in continuous unidirectional fibre-reinforced composites, where the tensile modulus is primarily dictated by the fibre content.

The observed results are compared with some relevant papers as given below. It is, however, essential to acknowledge the intricacies involved in comparing our research to existing studies, as the properties of composites are influenced by numerous variables. These variables include the glass fibre length and type (recycled or virgin), matrix type and composition, batch-to-batch variation (if applicable), manufacturing method, defects in the composites like voids and the quality of interfacial bonding. Despite these differences, a comparative analysis can shed some light on the usefulness of waste or recycled materials in comparison to virgin ones.

Sadik et al. [13] observed a similar trend whilst investigating extruded wGF-reinforced rHDPE composites, where both materials were ground to a powder before extrusion. A decrease in tensile modulus was observed as the GF content was increased from 0 – 30 wt%. This was reported to be due to imperfect adhesion between the polymer matrix and glass particles. An increasing amount of aggregation of glass particles was reported as the wGF content increased, making them unable to support stress transferred from the matrix. A tensile modulus of 0.69 – 0.56 GPa (0 wt% - 30 wt% wGF) was reported in that work compared to the 0.69 – 4.7 GPa achieved in this current investigation for the laminate with a similar wGF content (0 – 33 wt% wGF).

Abd Rahman's study [37], investigating short vGF (length 6 mm) reinforced vPP extruded composites, exhibited a tensile modulus of 4.8 GPa at 23 vol% vGF compared to 4.2 GPa in this current study (MD) at the same wGF content. It is important to note, however, that in this study the matrix is solely comprised of vPP, which has a higher unreinforced tensile modulus of 2 GPa, in addition to being a virgin material, whereas the tensile modulus of unreinforced wMP is only 0.69 GPa, due to its high LDPE content. The MD laminate from this study exhibited an increase of 517% compared to an increase of 110% in the literature study [37]. This comparative analysis highlights the significant advantages of short wGF-reinforced PE laminates over equivalent extruded/injection moulded ones.

Figure 8 shows the representative stress-strain curves of each of the laminates during tensile testing.

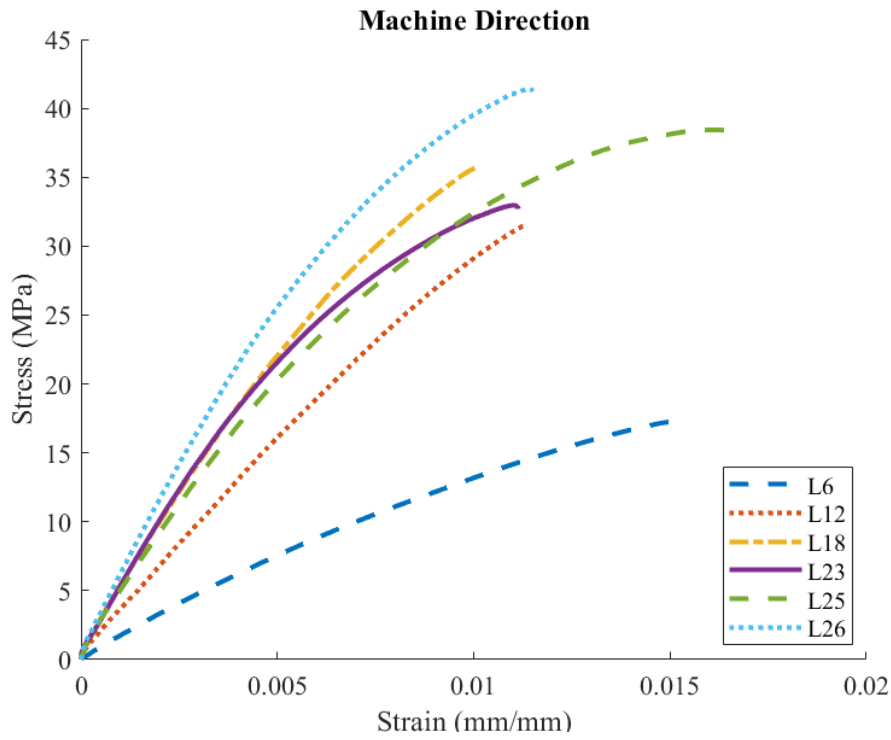


Figure 8a: Representative stress-strain curves from each waste mixed plastic (wMP)/waste glass fibre (wGF) laminate with samples cut in the machine direction (MD).

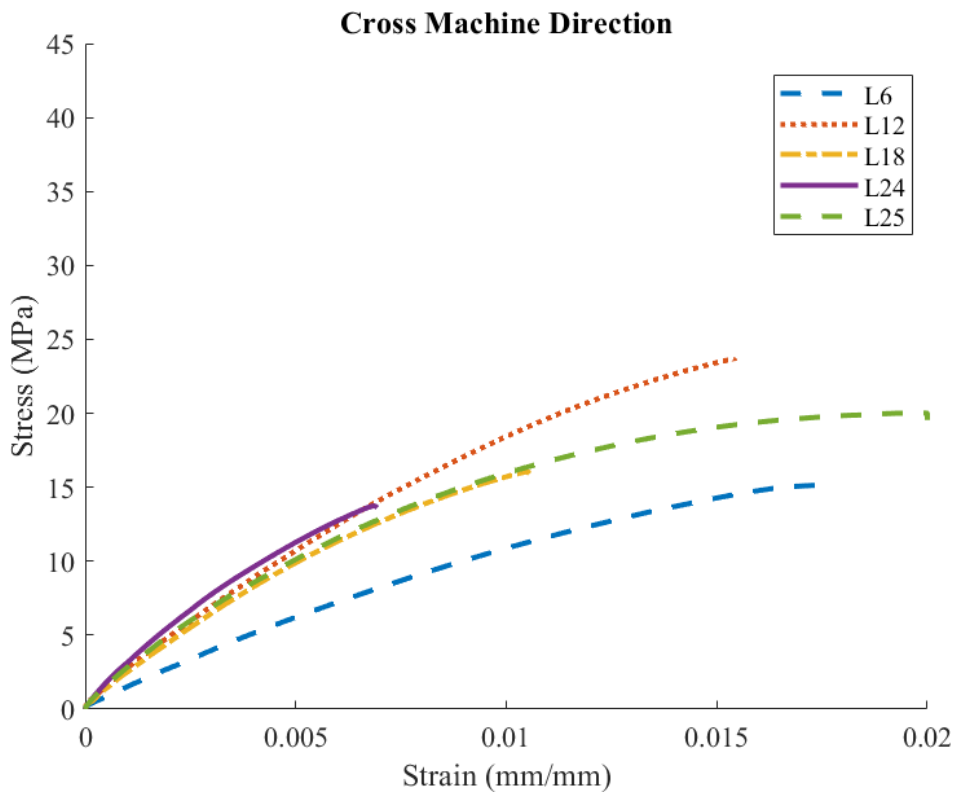


Figure 8b: Representative stress-strain curves from each waste mixed plastic (wMP)/waste glass fibre (wGF) laminate with samples cut in the cross machine direction (CMD).



As shown in Figure 8a, the stress-strain curves of MD laminates exhibit the expected behaviour, with the modulus and tensile strength generally increasing as the wGF content rises. Figure 8b, depicting CMD laminate results, shows stress-strain curves consistent with the average CMD outcomes. Notably, in both MD and CMD stress-strain curves (Figures 8a and b), higher wGF laminates demonstrate a more ductile behaviour before fracture, evident from the decrease in gradient preceding failure. This might be attributed to the increase in voids with the increase in wGF content in the laminates. The increased voids indicate the presence of poorly impregnated areas in the laminate. The lack of bonding with the fibres allowed the matrix to deform to a larger extent showing an overall higher ductility in the composites.

### 3.3.1 Theoretical tensile modulus of short fibre composites (with partial orientation) using the Halpin-Tsai equation.

The Halpin-Tsai equation was used to determine the theoretical tensile modulus of the wMP/wGF laminates. In the study conducted by Osoka [38], a modification to the Halpin-Tsai equation was proposed, enabling the calculation of the tensile modulus for discontinuous fibre composites with partially orientated fibres. The equation used was as follows:

$$E_c = \eta_l \eta_0 E_f V_f + E_m V_m \quad (6)$$

where:

$V_f$  = volume fraction of the reinforcement

$E_c$  = tensile modulus of the composite

$V_m$  = volume fraction of the matrix

$E_f$  = tensile modulus of the reinforcement

$\eta_0$  = fibre orientation distribution factor

$E_m$  = tensile modulus of the matrix

$\eta_l$  = packing efficiency factor

The packing efficiency factor, denoted as  $\eta_l$ , is an empirically derived term specific to the GF used, which can account for the void content in the resulting composite laminate. The fibre orientation distribution factor,  $\eta_0$ , is calculated using the following equation [39]:

$$\eta_0 = \sum_n a_n \cos 4 \theta_n \quad (7)$$

where  $a_n$  is the proportion of the fibres making an angle  $\theta_n$  to the MD. The  $a_n$  and  $\theta_n$  terms were obtained using several high-resolution images of the dry wGF, i.e. as-received wGF nonwoven mat, before extrusion coating, with the ImageJ software [40] using the OrientationJ plug-in, as shown in Figure 9. An average value of  $\eta_0$  was taken across 10 images each for both the MD and CMD.

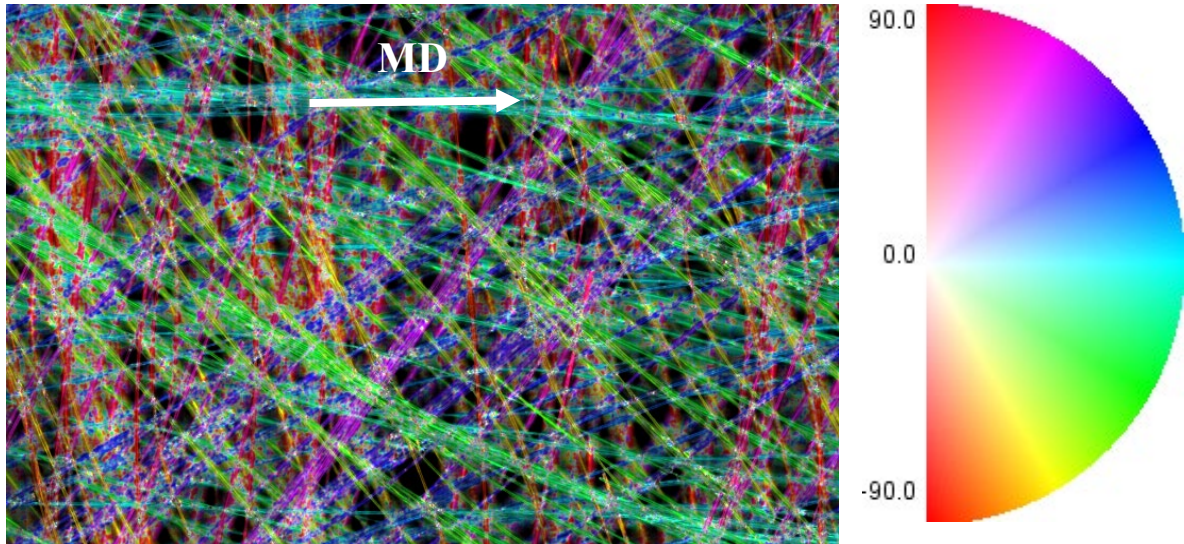


Figure 9: Image of the waste glass nonwoven mat (wGF) after processing using the OrientationJ plug-in.

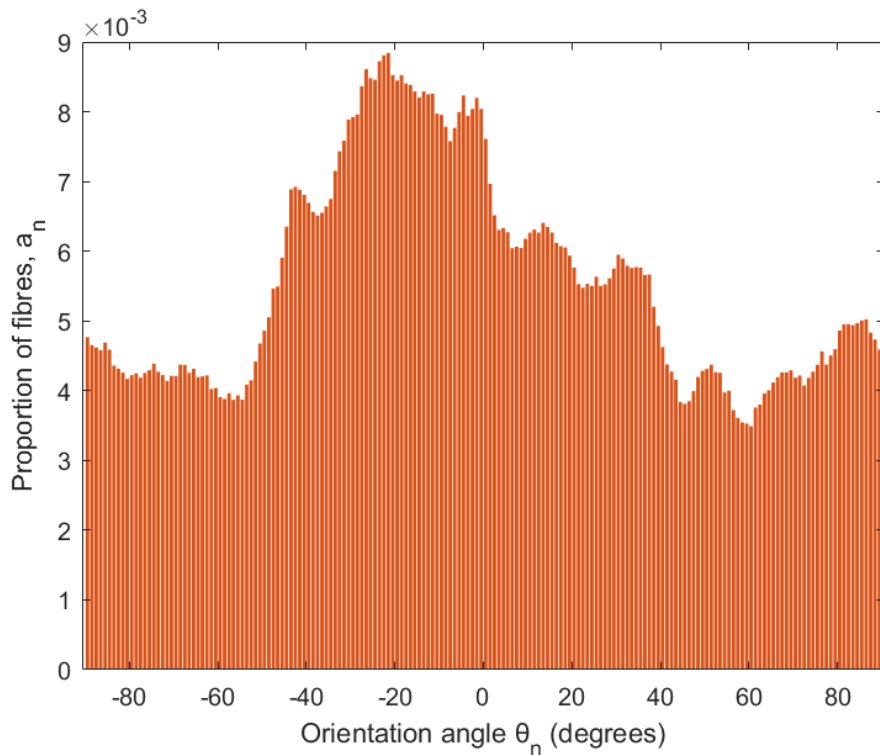


Figure 10: Graph showing the results output after a representative waste glass nonwoven mat (wGF) image has been processed using ImageJ with the OrientationJ plug-in.

A representative output of the OrientationJ software using one of the wGF images is shown in Figure 10. Using this method, an average value of  $\eta_0$  was calculated as 0.44 for the MD and 0.31 for the CMD. As the  $\eta_0$  value is higher for MD, this means that there are more fibres orientated in the MD than in the CMD direction. This is due to the manufacturing method of the GF mats, and the original fibre architecture in the mats; the extrusion coating process does not have an impact on the directionality of the fibres.

To determine the packing efficiency factor,  $\eta_l$ , the experimental tensile modulus values were substituted into the Halpin-Tsai equation (Eqn. 6) and corresponding  $\eta_l$  values were calculated for different wGF contents and plotted in Figure 11. An  $\eta_l$  value corresponding to a void content of 0 vol% was derived by employing a regression analysis, as illustrated in Figure 11.

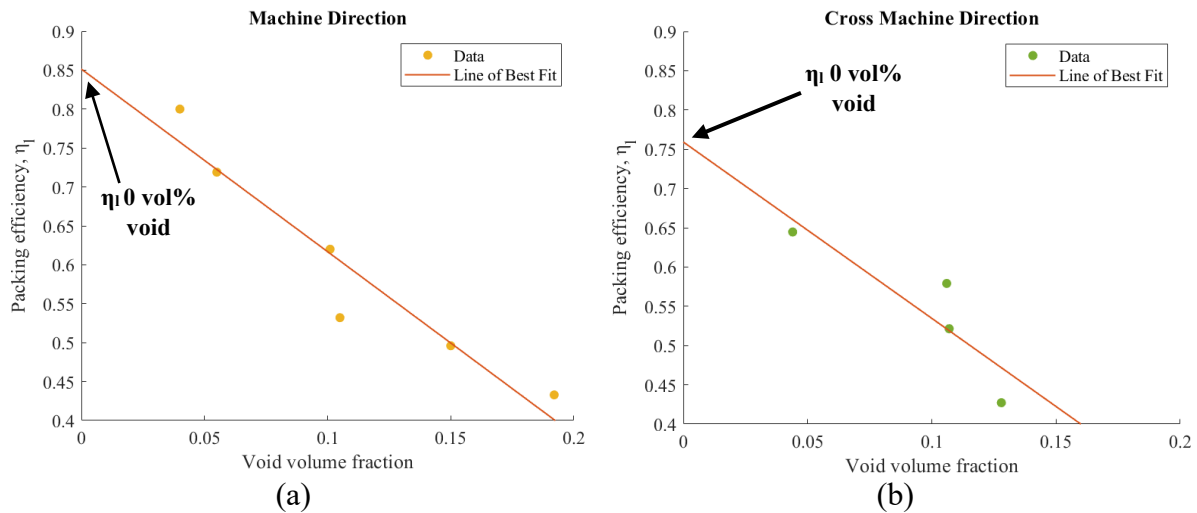


Figure 11: Plot of void content against packing efficiency  $\eta_l$  with line of best fit for (a) machine direction (MD) and (b) cross machine direction (CMD) cut samples.

Using the data presented in Figures 11a and b,  $\eta_l$  values of 0.85 and 0.76 were obtained for MD and CMD respectively corresponding to 0% void.

Looking at Figure 11, it is evident that there is a distinct  $\eta_l$  value for both MD and CMD. Thus, this packing efficiency factor  $\eta_l$  comprehensively captures the efficacy of fibre arrangements in the 2-D plies, as well as the voids in the interply and intraply regions. Therefore, it offers an insight into the effectiveness of the fibre compaction along the axis of the testing direction. This is different from the  $\eta_0$  value, which is used to modify the equation to account for the presence of non-aligned fibres in the testing direction.

Once the  $\eta_l$  values are obtained for 0% voids, the theoretical tensile modulus for wMP/wGF composite laminates is determined using Eqn. 6, as shown in Figure 12a. Figure 12b shows the theoretical tensile modulus values of wGF/wMP laminates with 10 vol% voids along with the experimentally measured tensile modulus values of wMP/wGF laminates containing approximately 10 vol% voids. The  $\eta_l$  values for 10% voids were obtained from Figure S3.

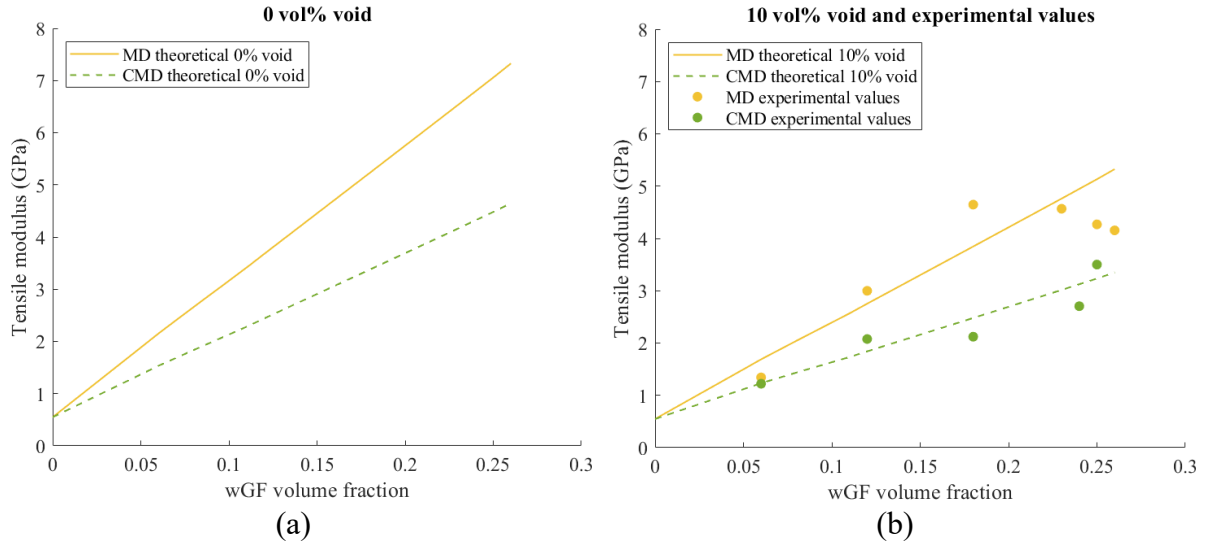


Figure 12: Graph showing the theoretical tensile modulus of waste mixed plastic (wMP) laminates reinforced with slightly directional short waste glass fibres (wGF), in the machine direction (MD) and cross machine direction (CMD) with (a) 0% voids and (b) with 10 vol% voids in comparison to the experimental values.

With improved quality of consolidation and lower void content, the experimental values are likely to match more accurately with the theoretically calculated values for the 0 vol% void content laminates (Figure 12a). In the case of the experimental MD samples, it becomes evident that laminates with higher volume fractions of wGF (L25 and L26) exhibit a lower tensile modulus than the theoretically calculated values (Figure 12b), which can be attributed to higher void contents in these laminates.

As the wGF content rises, achieving high-quality laminates with the chosen wMP coating thickness (0.95-1.31 mm) and the manufacturing pressure becomes more challenging since the wMP needs to impregnate numerous plies of dry wGF. One potential solution to this issue is to have thinner wMP coating on wGF that will allow a more uniform distribution of wMP across the thickness of the laminate during lay-up. This approach will reduce the number of dry wGF plies the wMP must penetrate during compression moulding and the packing efficiency is likely to be improved.

### 3.3.2 Theoretical tensile modulus of short fibre composites (with random orientation) using the Halpin-Tsai equation.

The Mori-Tanaka laminated analogy (MT-LA) approach was employed to calculate the theoretical tensile modulus of randomly oriented short fibre laminates. This encompasses both 2-D dispersion (laminated composites) and 3-D dispersion (extruded or injection-moulded composites) of short fibres. The objective was to quantify the difference in expected tensile modulus between randomly orientated laminates, partially orientated laminates - as investigated in this study - and extruded/injection-moulded composites. The Halpin Tsai equation for randomly orientated short fibre reinforced composites is as follows:

$$E_C = \alpha E_L + (1 - \alpha) E_T \quad (8)$$

where:

$$E_L = E_m \frac{1 + \frac{l}{r} \eta_L V_f}{1 - \eta_L V_f}, \quad E_T = \frac{1 + 2\eta_T V_f}{1 - \eta_T V_f} \quad (9)$$

$$\eta_L = \frac{\frac{E_r}{E_m} - 1}{\frac{E_r}{E_m} + \frac{l}{r}}, \quad \eta_T = \frac{\frac{E_r}{E_m} - 1}{\frac{E_r}{E_m} + 2}$$

where:

$E_C$  = Composite elastic modulus

$V_f$  = Volume fraction

$E_L$  = Longitudinal elastic modulus

$l$  = length of reinforcement

$E_T$  = Transverse elastic modulus

$r$  = radius of reinforcement

$E_m$  = Matrix elastic modulus

$\alpha$  = constant coefficient

$E_r$  = Reinforcement elastic modulus

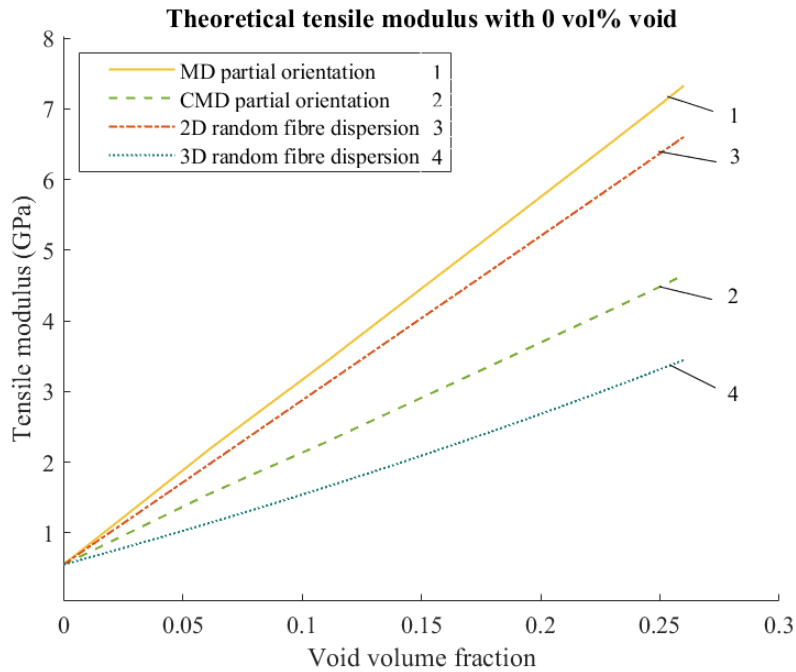
The MT-LA approach used for 2-D dispersion is given below [41]:

$$\alpha = 0.339 - 0.035V_f - 0.642 \left( \frac{E_m}{E_r} \right) \quad (10)$$

A different  $\alpha$  value is used in the case of 3-D dispersion:

$$\alpha = 0.13 + 0.0815V_f - 1.669 \left( \frac{E_m}{E_r} \right) \quad (11)$$

Using eqn. 10 and 11, it becomes possible to compute the theoretical variation in tensile modulus of composites with 2-D and 3-D dispersion of short fibres, all other factors remaining the same, as depicted in Figure 13.

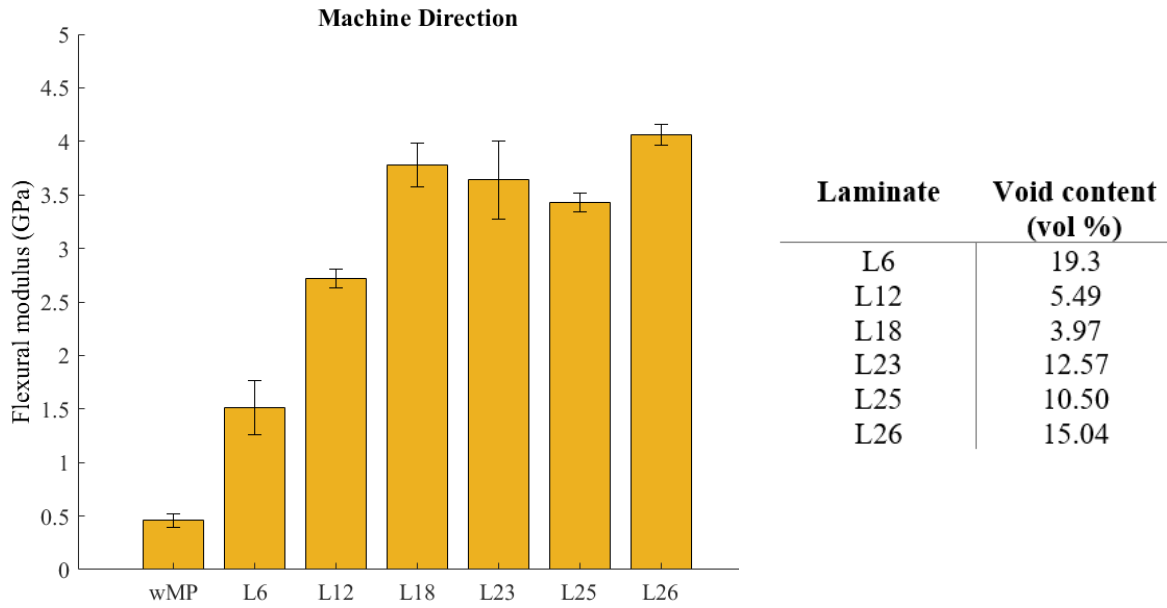


**Figure 13:** Graph comparing the theoretical tensile modulus of waste glass fibre (wGF) reinforced waste mixed plastic (wMP) laminates (containing 0 vol% voids) using different orientations of reinforcement.

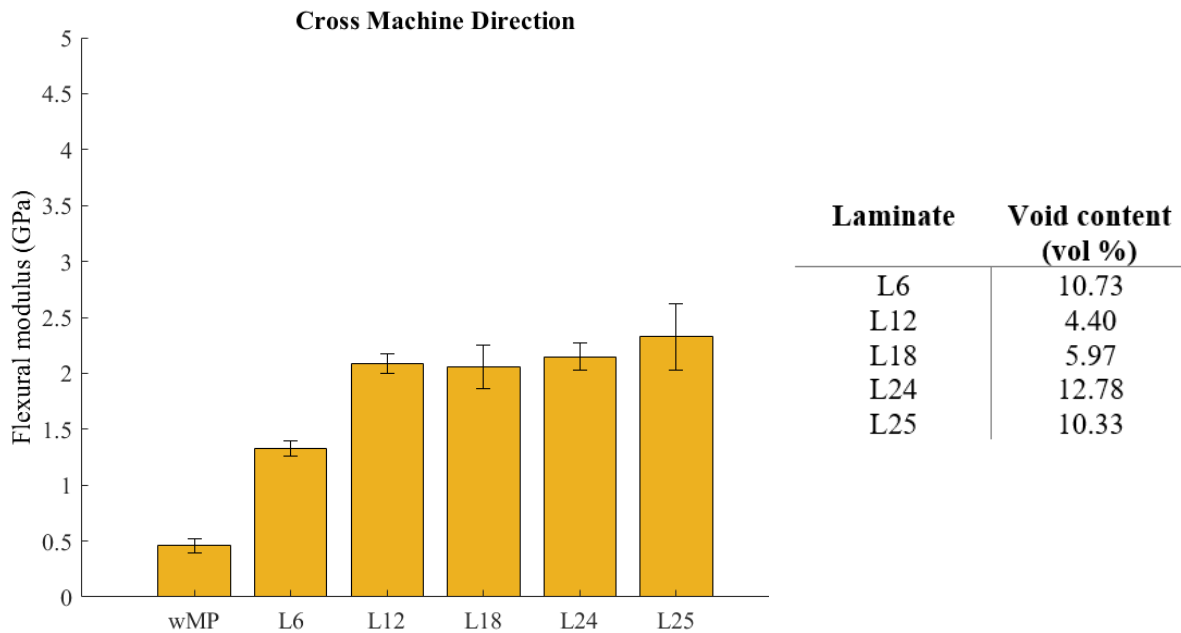
The findings indicate that regardless of whether the samples are cut in the MD, CMD, or randomly oriented, composites featuring a 2-D dispersion of short fibres (laminates) exhibit a higher tensile modulus compared to extruded composites, which display a 3-D dispersion of short fibres. This is a key finding and can guide the manufacturing of high-value composites using short waste fibres.

### 3.4 Flexural testing

The flexural test results of the wMP/wGF laminates are given below in **Figures 14a and 14b.**



**Figure 14a:** Comparison of the flexural modulus between unreinforced waste mixed plastic (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the machine direction (MD).



**Figure 14b:** Comparison of the flexural modulus between unreinforced waste mixed plastics (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the cross machine direction (CMD).

Similar to the tensile modulus results, the MD laminates exhibited an increase in flexural modulus values as the wGF content increased from 0 – 18 vol%, shown in **Figure 14a**. With a further increase in wGF content, the void content also increased which reduced the flexural modulus. L18, with a lower void content, showed a higher flexural modulus value than L23 and L25. The higher number of voids played a more dominant role in failure than the increase

in wGF content. A similar trend was observed with the CMD laminates (Figure 14b). The flexural modulus values did not show any significant increase in L24 and L25 due to high void content.

Huang et al. [27] reported vHDPE having a 582% increase in flexural modulus when reinforced with 40 wt% of short vGF (4 mm length). The flexural modulus changed from 0.85 GPa to 5.8 GPa. In the current study, the flexural modulus of wMP changed from 0.5 GPa to 3.6 GPa with 43 wt% of wGF, showing an increase of 620%. The differences in the starting material types, manufacturing methods and void contents need to be considered, as mentioned before in the case of tensile modulus.

### 3.5 Compression testing

Figures 15a and 15b show the compression strength results for all laminates with samples cut in the MD and CMD alongside unreinforced wMP.

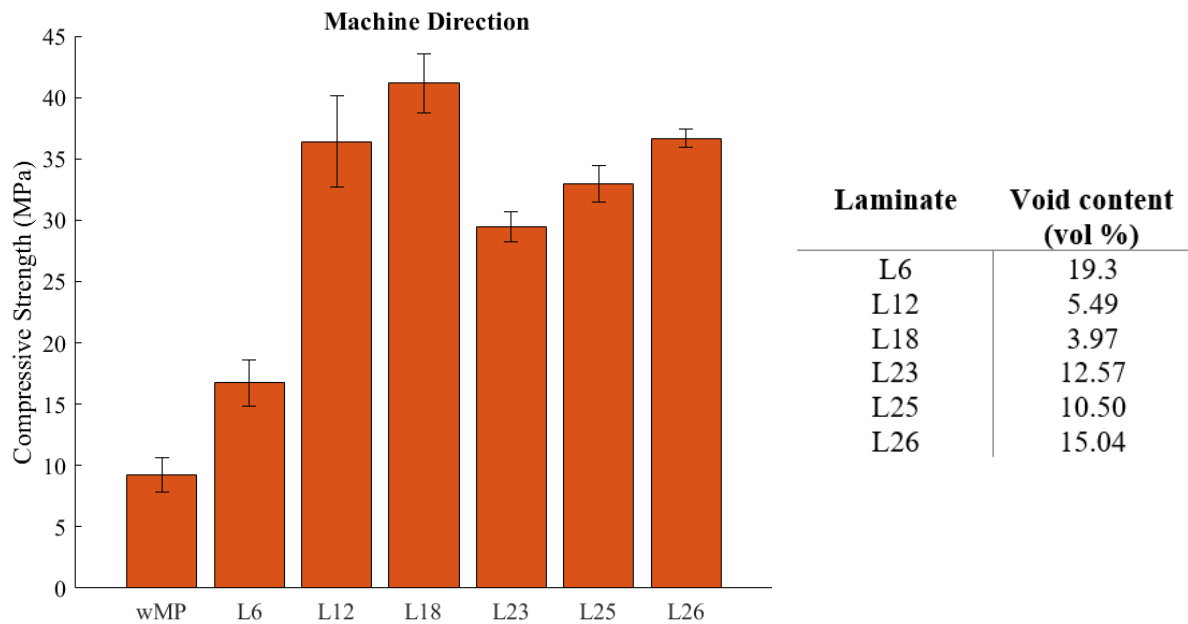
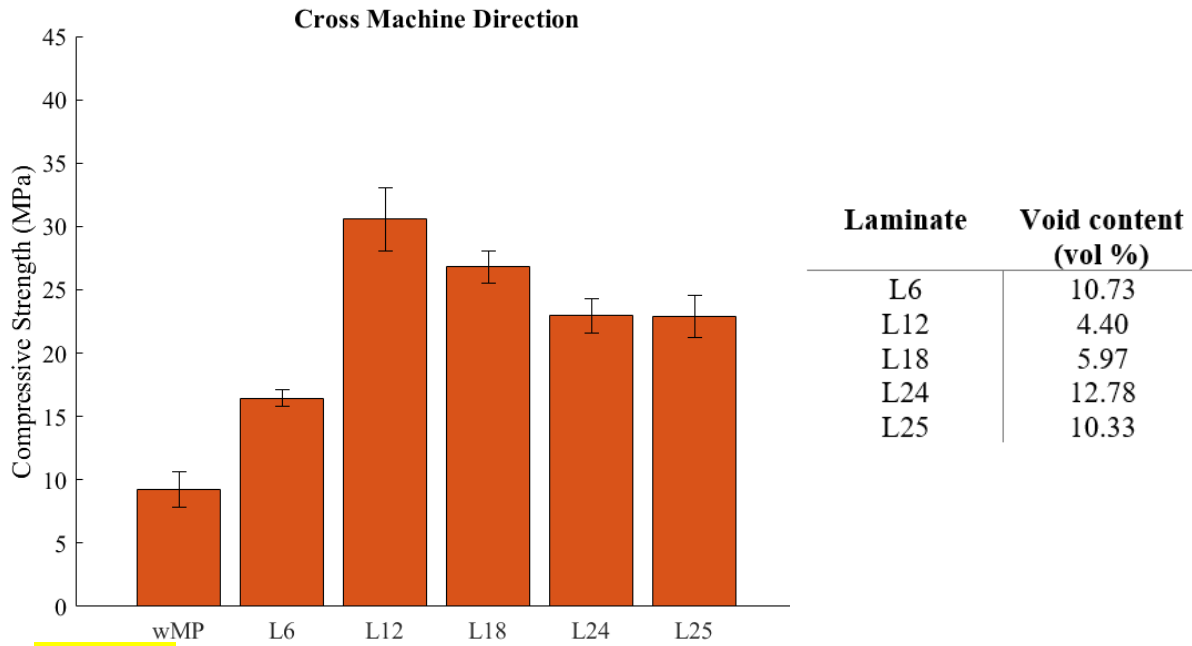


Figure 15a: Comparison of the compressive strength between unreinforced waste mixed plastics (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the machine direction (MD).

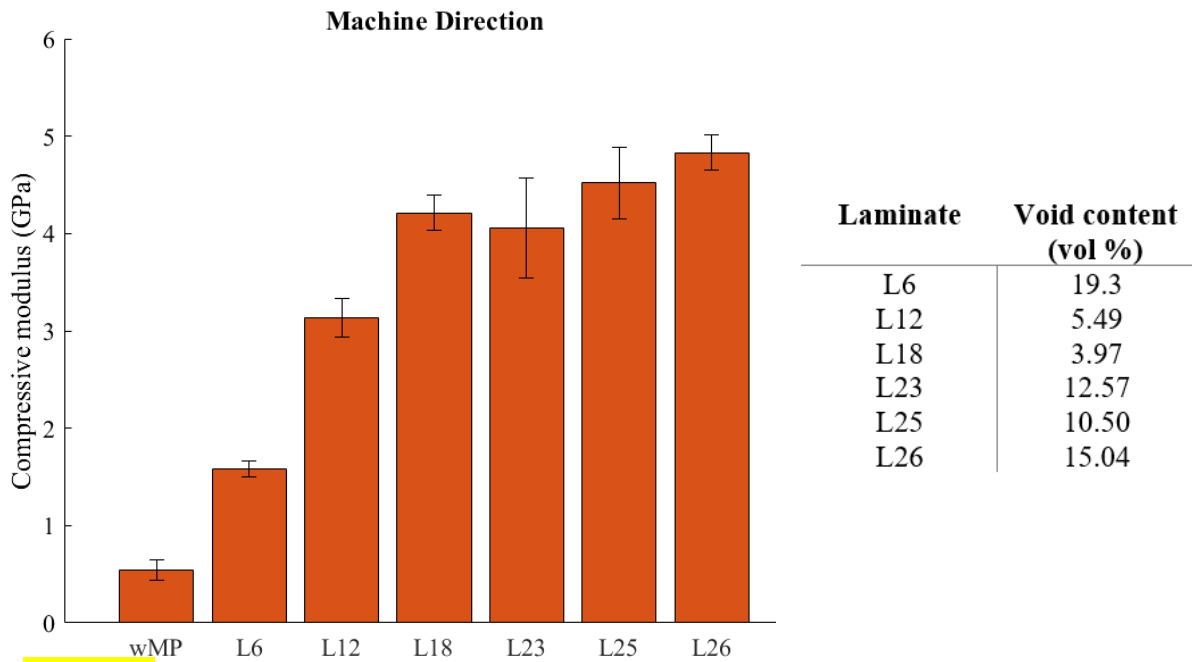




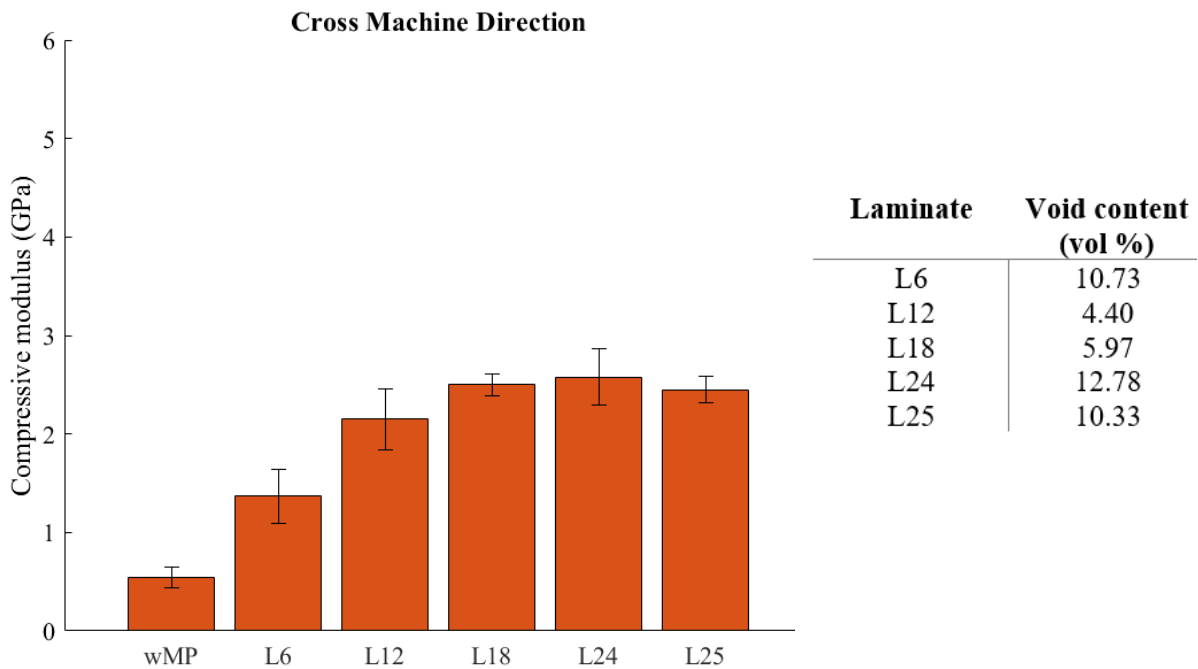
**Figure 15b:** Comparison of the compressive strength between unreinforced waste mixed plastic (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the cross machine direction (CMD).

**Figures 15a** and **b** show the compressive strength results of the wMP/wGF laminates in the MD and CMD respectively. The compressive strengths exhibit a strong correlation with the void content within the laminates. This is evident, particularly with MD laminates L12 and L18, as the compressive strength seems to increase proportionally with increasing wGF content and lower void content. There is a significant drop when the void contents rose above 10 vol% in L23, L25, and L26 (**Figure 15a**). An interesting point to note here is that, regardless of a void content of 15 vol%, the compressive strength of the L26 MD laminate is 296% higher than unreinforced wMP, and 106% higher than that of unreinforced virgin HDPE [30]. The observations were similar in the case of the CMD laminates, shown in **Figure 15b**. L12, with the minimum void content, exhibited the highest compressive strength. A strong correlation was evident between the void content and compressive strength and that superseded the effect of increasing wGF content in the laminates. The voids act as local stress concentrators leading to easier buckling or deformation of the composite, lowering the strength.

The current study revealed a compressive strength of 42 MPa for the MD laminate containing 18 vol% wGF, whilst a compressive strength of approximately 37 MPa was reported in an injection moulded wMP/vGF composite containing a similar GF content (12.7 vol%) [9]. This indicates the advantage of the 2-D dispersion of wGF within the composite, especially considering that the wMP mix used in [9] is likely to contain a significantly higher proportion of HDPE than in this current study, as the reported compressive strength value of the unreinforced wMP was comparable to vHDPE (19.7 MPa) [42].



**Figure 16a:** Comparison of the compressive modulus between unreinforced waste mixed plastics (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) cut in the machine direction (MD).



**Figure 16b:** Comparison of the compressive modulus between unreinforced waste mixed plastics (wMP) and waste mixed plastic (wMP)/waste glass fibre (wGF) laminates cut in the cross machine direction (CMD).

The compressive modulus of the samples increases as the wGF content increases as shown in **Figures 16a** and b. However, the compressive modulus values are less affected by the increase in void content as evident in L23, L25 and L26 than that observed in the case of compressive strength. This implies that, although voids introduce discontinuity in the matrix leading to localised areas of reduced stiffness, their effects are more pronounced in load-bearing ability under compression and that causes a bigger drop in compressive strength.

As shown in **Figures 16a** and b, compressive modulus values of 3.7 GPa and 2.3 GPa were obtained for the MD and CMD laminates respectively, with 12 vol% wGF content, while a compressive modulus of approximately 2.5 GPa was reported with a similar GF content (12.7 vol%) in [9]. Considering the unknown quantities of polyolefins within each investigation, it is worth noting that the compressive modulus increased by 141% in [9], while in this study, the compressive modulus increased by 480% in the MD, and 360% in the CMD.

Table 3 below shows a summary of the mechanical properties of wMP/wGF laminates with samples cut in the MD compared to wMP/vGF composites from the literature [9], with both composites containing similar GF contents (approximately 12 vol%), although the composites have different processing methods, void contents and type of GF reinforcement.

*Table 3: Table comparing the L12 MD laminate to a literature study containing similar composite materials [9].*

Material Comparison		Tensile		Flexural	Compression	
		Strength (MPa)	Modulus (GPa)	Modulus (GPa)	Strength (MPa)	Modulus (GPa)
<b><u>Current study</u></b> <b>GF:</b> Waste non-woven mat, 18mm length, 13µm diameter. <b>Matrix:</b> wMP consisting of majority LDPE. <b>Processing:</b> Compression moulding. <b>wGF content:</b> 12 vol%. <b>Void content:</b> 5.5 vol%	Value	28.0	3.00	2.71	36.4	3.70
	% Increase from un-reinforced	188%	332%	490%	293%	480%
<b><u>Bajrachayra [9]</u></b> <b>GF:</b> Virgin chopped GF, 4mm length, 13µm diameter. <b>Matrix:</b> wMP consisting of unknown quantities of waste HDPE, LDPE, and PP. <b>Processing:</b> Injection moulding <b>vGF content:</b> 12.7 vol%. <b>Void content:</b> Unreported	Value	25.7	3.07	3.30	36.7	2.48
	% Increase from un-reinforced	74%	240%	357%	85%	141%

Table 3 shows that, when compared to an injection moulded composite containing similar materials, the mechanical properties are quite similar. The percentage increase in the current study is significantly higher than the increase in the referenced study, particularly the compressive properties. While there are some differences in the material types, this highlights the possible benefits of a laminated manufacturing approach.

### 3.6 Fracture surface analysis

Figure 17 shows SEM micrographs of tensile fracture surfaces of the laminates, revealing the quality of interfacial bonding between the wMP and wGF.

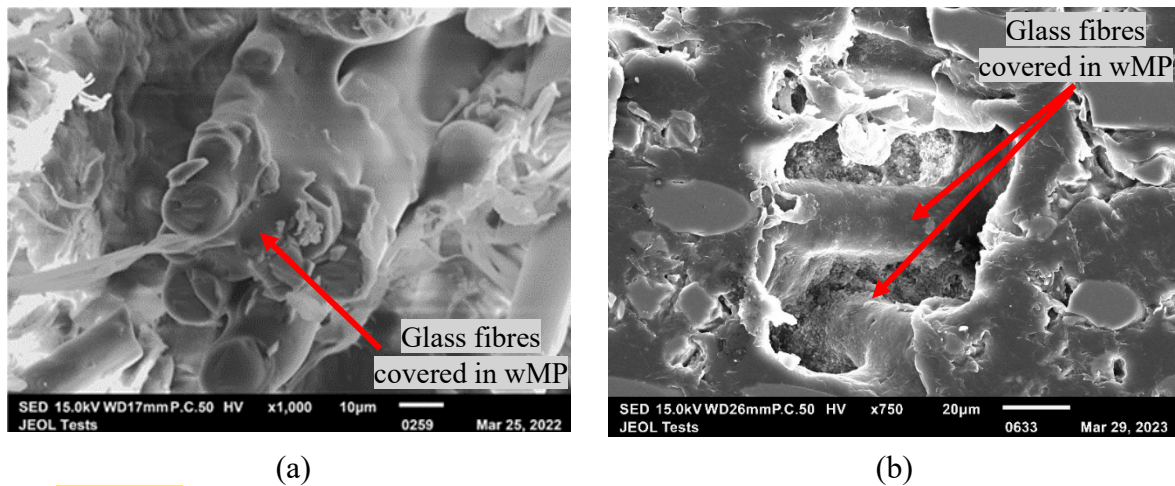
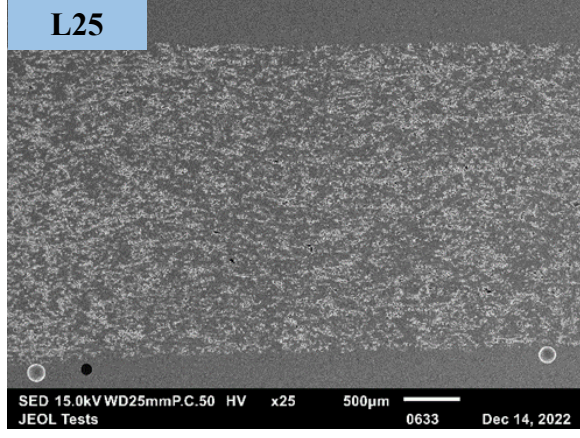
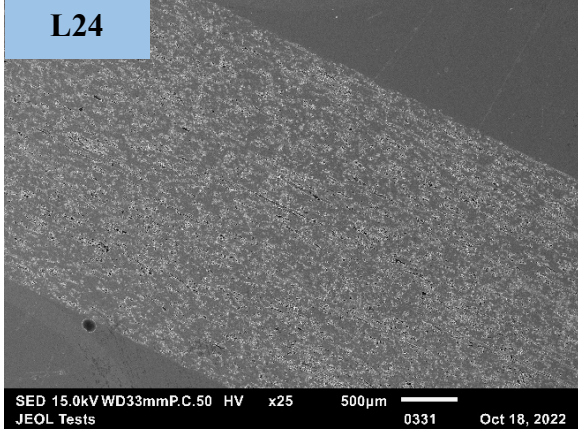
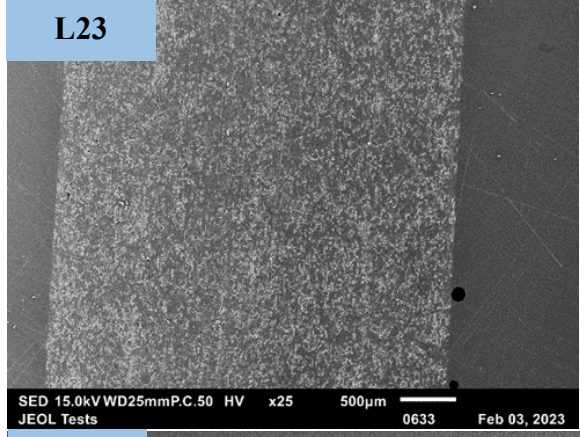
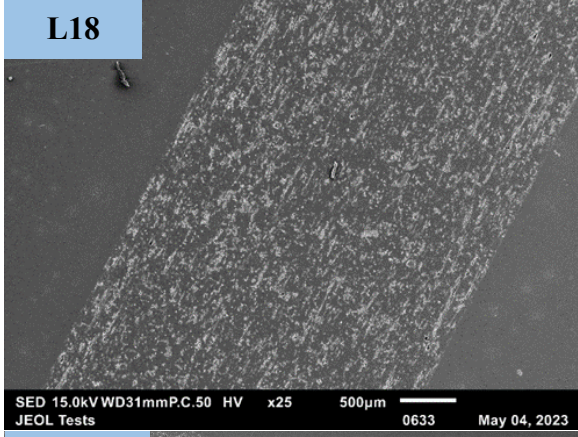
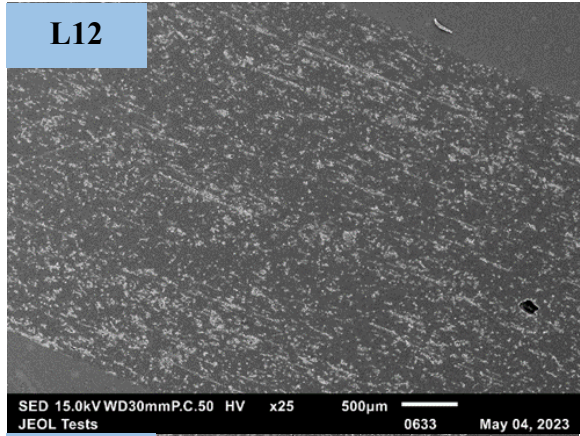
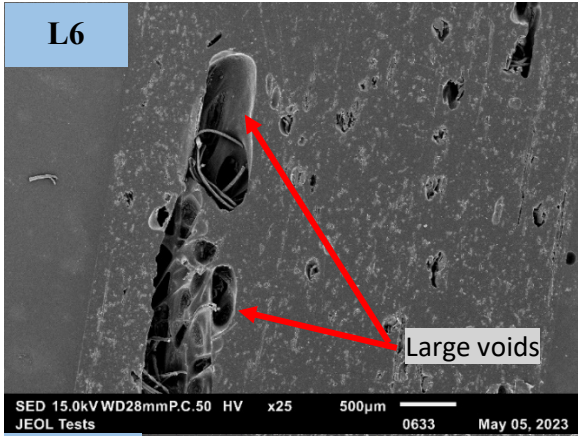


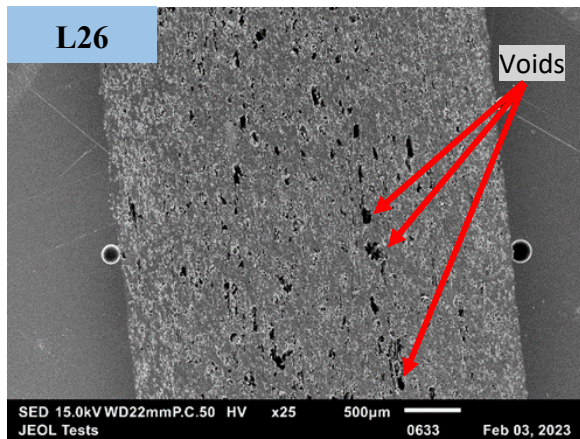
Figure 17: SEM image of (a) a tensile fracture surface and (b) a cross-section, showing the interface between waste mixed plastics (wMP) and waste glass fibre (wGF) in the 23 vol% machine direction (MD) cut laminate.

A well-covered wMP/wGF interface is evident in the micrographs (Figure 17), indicating that the presence of the urea formaldehyde binder on the wGF worked effectively as a sizing agent for wMP/wGF composites. This was the case in all the laminates with different wGF contents, shown in Figure S4.

The cross sections of wMP/wGF laminates with different wGF contents were examined under SEM to assess the quality of impregnation and consolidation. Representative SEM images were taken for each wGF content as shown in Figure 18.







**Figure 18:** SEM images of cross sections at  $\times 25$  magnification showing the impregnation of the waste mixed plastic (wMP) across the thickness of the laminates with different waste glass fibre (wGF) contents.

Examining the L6 laminate depicted in **Figure 18** reveals the presence of large voids, which aligns with the elevated void contents measured within both MD and CMD L6 laminates, particularly in the MD. Similarly, L26 exhibited a heightened void content of 15 vol%, but the voids are much smaller and with higher distribution throughout the cross section.

#### 4. Conclusions

This study investigated the manufacturing and characterisation of waste mixed plastics (wMP) based composites reinforced with waste glass fibres (wGF). wMP from plastic film packaging waste, separated by float-sink method, was effectively utilised in combination with wGF nonwoven mat to produce value-added composites. The amount of GF in the machine direction (MD) and cross machine direction (CMD) of the mat were different and that had a significant effect on the mechanical properties. Thermoplastic prepregs produced with wGF and wMP were highly beneficial for manufacturing consistent quality laminates. The composites were manufactured by compression moulding process and exhibited voids between 4-19 vol%.

The tensile strength and modulus increased by up to 330% and 570% respectively compared to the unreinforced wMP. The tensile modulus of both MD and CMD samples demonstrated a substantial increase compared to the theoretically calculated values for extruded composites. This indicated the benefits of laminates incorporating wGF nonwoven mats with 2-D dispersion of fibres over extruded composites with 3-D dispersion of fibres made from the same materials. The flexural modulus of the laminates increased up to 780% compared to unreinforced wMP. A significant improvement was also observed for the compressive properties. The compressive strength increased up to 350% and 230% for the MD and CMD laminates respectively, while the increase in compressive modulus was 700% and 400% for the MD and CMD laminates respectively. It was evident that the presence of voids had a more detrimental effect on the compressive strength than on the compressive modulus.

The consolidation quality, packing efficiency and optimum loading of wGF in the manufactured laminates were dependent on factors like the thickness of the wMP coating on the wGF ply in the prepreg, consolidation pressure and fibre orientation in the wGF mat. In

this study, 12-18 vol% of wGF content was found to be optimum leading to minimum voids and the best combination of properties.

These findings demonstrate the substantial enhancements in mechanical properties that can be achieved by utilising wMP and wGF in laminates, highlighting their potential for diverse applications. Addressing void content becomes a key focus for unleashing the full potential of these composites. This scientific comprehension carries profound implications for the circular economy, showcasing the enhanced mechanical properties attainable by combining low-cost packaging waste with waste reinforcement. Additionally, the potential increase in mechanical properties through laminate manufacturing over extrusion offers insights into effective manufacturing routes for higher-value products from waste.

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