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IMPACT OF LIGNIN FILLER ON MECHANICAL PROPERTIES AND ANISOTROPIC BEHAVIOR OF COMPRESSION MOULDED GLASS/POLYPROPYLENE LONG FIBRE THERMOPLASTICS

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***Abstract*—** Glass Long Fibre Thermoplastics (LFTs) are becoming increasingly popular as lightweight, high-performance recyclable materials. This trend has also led to greater interest in the use of biomaterials, such as bio-fillers like lignin, to help reduce the carbon footprint of petroleum-based polymer composites. To evaluate the impact of the bio-filler (Kraft lignin) on the mechanical properties of glass-reinforced polypropylene (glass/PP) LFTs, specimens were extracted from 4 mm flat plaques manufactured through a compression moulding process. The study examined three different weight percentages of lignin: 0%, 14%, and 21% while keeping the glass fibre content constant at 30% by weight. Quasi-static tensile and shear tests were conducted to analyse the mechanical properties of the material. Additionally, in order to investigate anisotropy, samples were tested in two material orientations, namely, 0° and 90° material directions which will be precisely defined in this paper. Specimens from both the charge and flow regions were tested to assess their effect on the mechanical properties of the LFT material. The findings revealed that tensile strength decreased with increasing lignin content for the 0° orientation. Specifically, at 21% lignin content, the reduction in strength was notably higher at 36%, compared to just a 15% decrease observed with 14% lignin content. In the 90° material direction, a slight increase in tensile strength was noted for the 14% lignin content when compared to the 0% lignin condition. In the 0° orientation, strength consistently reached up to 100 MPa for samples from both the charge and flow regions, whereas the 90° orientation exhibited tensile strengths of up to 43 MPa. In the 0° orientation, samples from the flow region demonstrated higher tensile strength than those from the charge region. Conversely, in the 90° orientation, the charge region samples surpassed the flow region samples in strength by approximately 66% for the 0% lignin condition. The influence of the filler material on shear strength was found to be minimal for 14% lignin specimens and a slightly higher reduction of approximately 10% - 15% in the case of 21% lignin specimens. This study provides essential material characterisation, enabling more accurate numerical and analytical modelling of LFT composite materials.

Keywords- Long Fibre Thermoplastic; Biofillers; Lignin; Mechanical characterisation; Anisotropy

I. INTRODUCTION

In recent years, thermoplastic matrix composites have gained popularity due to their unique characteristics, including reheating capability, fracture toughness, improved impact resistance, recyclability, and cost-effectiveness compared to traditional thermoset materials and processes [1, 2]. While these composite materials offer numerous advantages over conventional counterparts, they often depend on non-renewable synthetic components, such as petroleum-based polymers. This reliance leads to two significant environmental issues: the depletion of finite petroleum resources and a lack of biodegradability. As a result, industry is increasingly focused on researching more sustainable materials [3, 4].

Composites incorporating bio-based fillers, such as lignin, can effectively balance industrial demands for mechanical integrity with the need for more sustainable materials. Lignin, one of the most abundant biopolymers on Earth, second only to cellulose, emerges as a strong candidate for use as a bio-filler in composites [5]. By incorporating bio-fillers such as lignin, the carbon footprint of polymers like polypropylene (PP) utilised as matrix compounds in composites can be significantly reduced.

Filler materials in long-fibre thermoplastic composites have traditionally been used to reduce costs, achieve desired dimensions, and enhance the mechanical properties or toughness of the composites [6]. Several studies have documented how different fillers influence the mechanical properties of composite materials. For instance, Kiran et al. found that small amounts of silicon carbide and titanium

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dioxide improved the mechanical properties of composites when used as filler materials. Additionally, calcium carbonate ($CaCO_3$) has been employed as a cost-reducing filler in the automotive industry and has also been shown to act as a toughening agent when used with coupling agents [7].

There have been few studies examining the influence of lignin on composites, primarily focusing on various polymer blends without fibre reinforcements. Sallem-Idrissi et al. [8] found that lower concentrations of lignin had no effect on the mechanical properties, while higher percentages of lignin resulted in a decrease in strength for polyamide 6 (PA6) and lignin blends. Kun et al. [9] reported a reduction in tensile strength with increasing content of Kraft lignin across various polymers such as polyethylene terephthalate (PET), polypropylene (PP), low-density polyethylene (LDPE), and polystyrene (PS). To enhance the mechanical properties of lignin-filled composites, Bozsódi et al. [10] investigated the role of a compatibiliser in improving interfacial adhesion in polypropylene/lignin blends.

LFTs manufactured through compression moulding are attracting considerable interest from the industry due to the benefits of this process compared to other methods, such as injection moulding [11]. LFT materials produced using compression moulding are sensitive to the charge and flow regions and demonstrate anisotropic characteristics, as documented by Bondy et al. [12] and Mohammadkhani et al. [13].

This study investigates the influence of lignin as a filler material on the mechanical properties of glass/polypropylene (PP) long-fibre thermoplastics. It explores the impact of lignin on the anisotropic behaviour and sensitivity of LFT to charge and flow regions. Additionally, the research examines how lignin affects the strain rate dependency under tensile loading, aiming to derive fundamental material properties for use in analytical and numerical modelling frameworks.

II. Materials and Methods

A. LFT-D Process and Materials

The direct compounded compression moulding process has generated significant interest as a manufacturing method for long fibre-reinforced thermoplastic materials due to its efficiency in producing high-strength structural parts [11]. In this study, LFT specimens examined were made with lignin-infused glass fibre reinforced/polypropylene (PP) manufactured at the Fraunhofer Innovation Platform for Composites Research at Western University in London, Ontario.

The process, as shown in Fig. 1, consists of two main stages, namely, compounding and moulding. During the compounding stage, polypropylene granules and lignin were fed into a compounding extruder (ZSE) operating at 230°C and 78.5 rpm. In this extruder, the granules were melted and mixed with lignin. The molten mixture then passed through a waterfall die, where glass fibres were introduced. This combined material was then fed into a chopper to cut the fibres to the desired length. Subsequently, the chopped molten material was introduced to a second corotating twin-screw

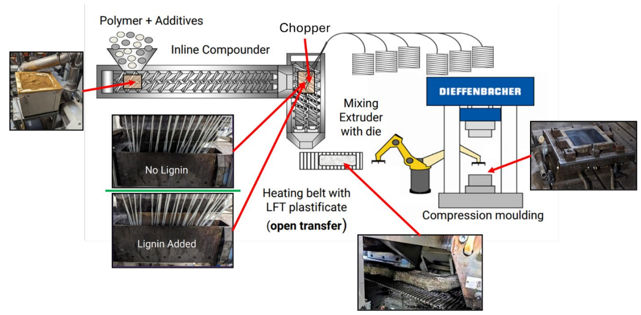

Figure 1. Glass/PP (lignin) direct compounded compression moulding process at FIP.

extruder (ZSG), also operating at 230°C but at 21.2 rpm. This step ensured the homogenisation of the fibres with the molten polymer and lignin mixture.

During the compression moulding stage, the plastificate (charge) obtained from the compounding process was placed into the Dieffenbacher DCP-U 2500/2200 press. The press was equipped with a flat plaque mould measuring 457 mm by 457 mm, which produced flat plaques with a nominal thickness of 4 mm. The plasticised material was pressed at a pressure of 150 bar for 60 seconds, followed by a 60-second cooling period.

The filler material used in this study was Kraft lignin, supplied by West Fraser. To reduce its moisture content from 40% to approximately 10%, the lignin was air-dried for seven days in a climate-controlled laboratory at room temperature. Three different variations of glass/polypropylene (PP) lignin-filled composites (LFTs) were produced to analyse the effect of lignin content on the mechanical properties of the LFT material. In all variations, the fibre content was kept constant at 30% by weight. The three different lignin filler ratios tested were 0%, 14%, and 21%, with the remaining percentage made up of polypropylene in each variant.

B. Sample Preparation

Specimens for both uniaxial tension tests and shear tests were extracted from a flat plaque using waterjet cutting. The

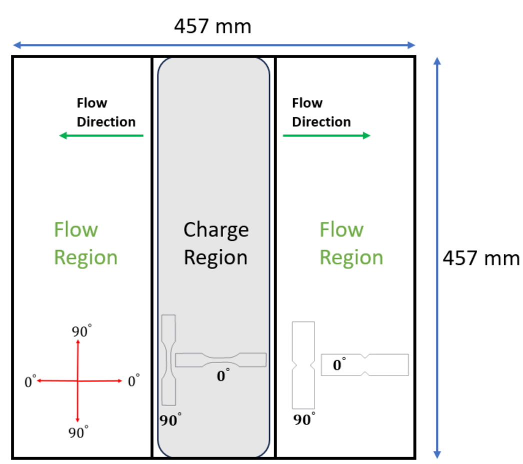

Figure 2. LFT plaque depicting various regions and orientations for specimens.

compression moulding process creates two distinct regions within the plaque: the charge region, where the molten material is initially placed, and the flow region, where the molten material moves within the mould during compression. Specimens were obtained from both regions for each test due to the varying material characteristics in these two areas. The direction in which the material flows during compression moulding is referred to as the material direction; this is also the direction in which the majority of the fibres align or orient themselves, known as the fibre orientation direction. Based on the material direction, as presented in Figure 2, samples with 0° and 90° directions were analysed for both tests in order to analyse the anisotropy of the material.

III. Experimental Methods

A. Uniaxial Tensile Testing

Quasi-static tensile tests were conducted utilising a 50kN capacity MTS Criterion (model 43) electromechanical universal testing machine at orientations of 0° and 90° with respect to the flow direction for all three variants of the materials with different lignin weight percentages. An MTS video extensometer equipped with a Nano-M1450 camera sampling at 5 Hz was utilised for strain measurement within the gauge region. Tensile tests were conducted at an approximate strain rate of 0.0001 s^{-1}, achieved with a crosshead speed of 0.001 mm/s. The ASTM D638 Type V specimen geometry was utilised for the tensile test samples.

B. V-Notched Beam Shear Testing

The V-Notched Beam Shear testing apparatus (Iosipescu) was mounted on the MTS machine and was utilised for quasi-static shear tests. For the shear tests, the crosshead speed of 0.01 mm/s was applied and maintained in order to achieve an approximate strain rate of 0.001 s^{-1}. The ASTM D5379 standard was utilised for the specimen geometry for the shear tests.

For optical strain measurement, an MTS video extensometer equipped with a Nano-M1450 monochrome camera (1088 x 1456 pixels) was utilised, sampling at 10Hz. The engineering shear strain was calculated within the gauge region as per ASTM D5379 using (1):

$$\varepsilon_e = |\,\varepsilon_{45°}\,| + |\,\varepsilon_{-45°}\,| \quad (1)$$

In equation (1), ε_e represents the engineering shear strain obtained by summing the strain magnitudes in the 45° ($\varepsilon_{45°}$) and -45° ($\varepsilon_{-45°}$) directions within the notch region of the shear samples.

IV. Results and Discussions

To analyse the impact of lignin, the bio-filler, on the mechanical properties of glass fibre-reinforced polypropylene LFT, uniaxial tensile tests and V-notch shear beam tests were performed. It is essential to characterise this thermoplastic composite in both the flow and charge regions due to the differences in mechanical properties exhibited in these areas. Consequently, samples from both regions were examined, and in both the 0° and 90° material directions, to assess their anisotropic behaviour. As previously indicated, fibre content remained constant at 30% by weight for all the variants with different lignin content. Correspondingly, with an increase in lignin, the polymer material content was reduced.

A. Stress-Strain Response for Tensile and Shear Tests

Fig. 3 illustrates the engineering stress-strain response for the samples obtained from both charge and flow regions for all three variations of lignin filler with 0° material direction. The mechanical strength of the 0% lignin content samples, did not exhibit any significant difference (less than 10%) in the tensile strength for the samples obtained from the two regions, with even lower deviation observed for the specimens with 14% and 21% lignin content. A decrease in tensile strength and elongation to failure was observed with an increase in lignin content. In the case of specimens from the flow region with 21% lignin content, the tensile strength and elongation to failure were significantly reduced compared to the previous two variants with 0% and 14% lignin content.

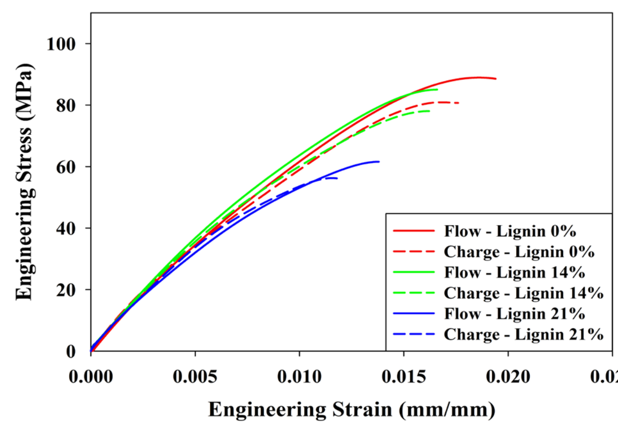

Figure 3. Tensile test engineering stress-strain responses for specimens with 0° material direction and varying lignin weight percentage from both flow and charge regions.

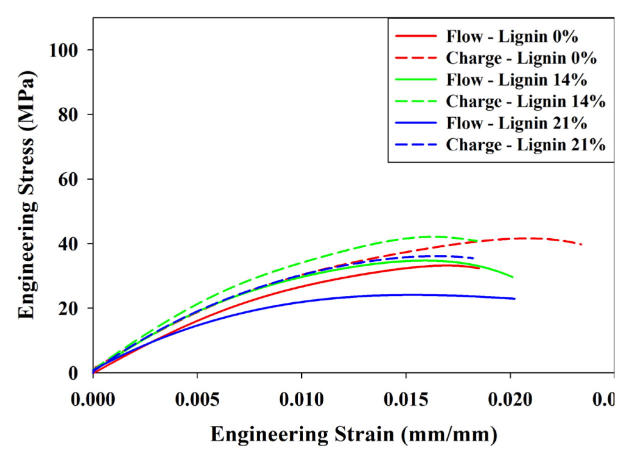

Figure 4. Tensile test engineering stress-strain response for specimens with 90° material direction and varying lignin weight percentage from both flow and charge regions.

Fig. 4 illustrates the engineering stress-strain response for the 90° material direction of samples taken from both the flow and charge regions, varying in lignin content. The samples from the charge region exhibited higher tensile strength across all material configurations. This difference in behaviour between the 0° and 90° material directions can be attributed to the restrictive flow of fibres and molten material in the charge region. As a result, some fibres retain their original orientation, unlike in the flow region, where most fibres align with the flow direction. This phenomenon causes some of the fibres in the charge region to align with the 90° material direction rather than being perpendicular to it, thereby enhancing the strength of samples from the charge region compared to those from the flow region with the same material direction.

The tensile strength and elongation of samples with varying lignin content from the charge region showed little variation. In the flow region, the samples containing 0% and 14% lignin exhibited similar tensile strength. However, the samples with 21% lignin displayed a decrease in strength. Additionally, there was no significant difference in elongation to failure among samples from the flow region, regardless of their lignin content.

The tensile strength significantly decreased in the 90° material direction when compared to the 0° material direction, which is an expected outcome. In the 0° direction, the fibre orientation aligns with the loading direction, enhancing the material's strength due to the higher strength of the fibres relative to other components. Conversely, in the 90° direction, the fibres are oriented perpendicular to the loading direction, rendering them ineffective in supporting external loads. As a result, the entire load is borne by the matrix, which possesses a considerably lower strength compared to the glass fibres.

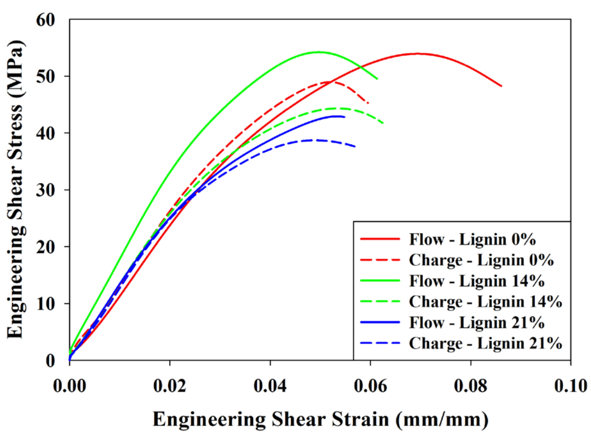

Figure 5. Shear test engineering stress-strain response for specimens with 0° material direction and varying lignin weight percentage from both flow and charge regions.

The engineering stress-strain responses for shear tests of specimens oriented in the 0° material direction are illustrated in Fig. 5. The samples taken from the flow region exhibited similar shear strength for both 0% and 14% lignin content; however, there were significant differences in strain to failure and shear modulus for samples from flow region. Specifically, the samples from the flow region with 14% lignin displayed lower strain to failure and a higher shear modulus. In both configurations, the flow region samples demonstrated greater shear strength compared to those from the charge region.

There was a notable decrease in shear strength for samples containing 21% lignin, regardless of whether they were from the flow or charge region. This increase in lignin content strongly impacted the shear strength at higher percentages.

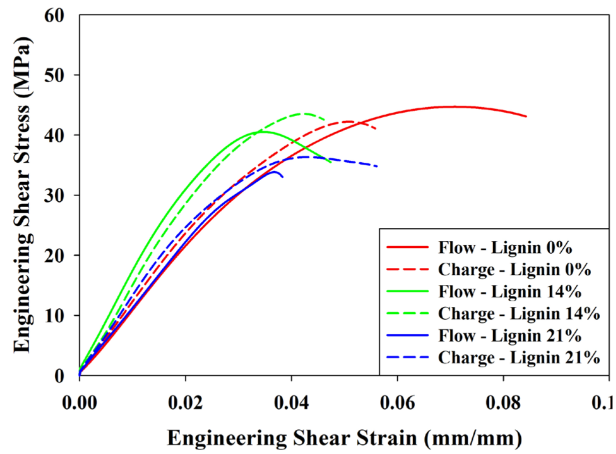

Figure 6. Shear test engineering stress-strain response for specimens with 90° material direction and varying lignin weight percentage from both flow and charge regions.

Interestingly, the engineering shear strain to failure remained largely consistent across various material configurations and regions, with one exception: the samples from the flow region with 0% lignin exhibited a higher strain to failure than all the others.

The shear stress-strain response for the 90° material direction is illustrated in Fig. 6, which includes samples from both the charge and flow regions. As noted in the earlier tensile tests, a similar trend is observed in the shear tests. Specifically, the samples from the charge region exhibited higher shear strength compared to those from the flow region, with the exception of samples that contained 0% lignin. In the case of 0% lignin, the shear strength of the flow region specimens was found to be slightly higher. When comparing the 90° material direction to the 0° material direction, a reduction in shear strength was observed for the 90° material direction, however, this decrease was not as significant as that was seen in the case of tensile strength.

B. *Influence on Mechanical Properties*

Fig. 7 details the influence of lignin, material direction and region from where the samples were extracted on the ultimate tensile strength (represented by bars) and Young's modulus (represented by lines). For 0° material direction from the flow region, the ultimate strength dropped from 100 MPa to 85.6 MPa as the lignin content increased from 0% to 14% and further dropped to 63.7 MPa with 21% lignin. The samples with 0% lignin from the charge region saw a significant drop compared to the corresponding sample from the flow region, but a minimal difference was observed for samples with 14%

and 21% lignin as compared to the corresponding samples from the flow region. Slight variation in Young's modulus was observed for samples with 0% and 14% lignin from both regions ranging from 7.3 GPa to 6.8 GPa, but for 21% lignin

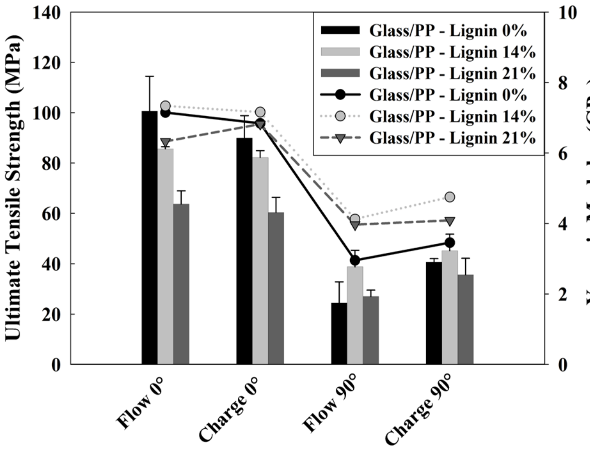

Figure 7. Ultimate tensile strength and Young's modulus for all material configurations.

content, an increase from 6.3 GPa to 6.8 GPa in modulus was observed between samples from flow region to charge region.

In the 90° material direction, the ultimate strength decreased from an average of 90 MPa to 40 MPa when compared to the 0° material direction. In this specific orientation, the samples containing 14% lignin content significantly outperformed the 0% lignin configuration, demonstrating an increase of approximately 58% for samples from the flow region and 12.5% for those from the charge region. Additionally, Young's modulus was found to be comparable for the 14% and 21% lignin samples from both regions, averaging around 4.2 GPa, while the 0% lignin samples exhibited a lower modulus, averaging 3.5 GPa.

Fig. 8 presents details on the ultimate shear strength (represented by bars) and shear modulus (represented by lines) of samples with varying lignin content and fibre orientation extracted from the flow and charge regions. The ultimate shear strength averaged around 44 MPa across different material configurations, with slightly lower values observed for samples oriented in the 90-degree material direction. Similar trends were observed for shear modulus averaging at 1.3 GPa for all material configurations.

V. Conclusion

This study examined how lignin filler content, material direction, and flow or charge regions affect the mechanical properties of glass/PP long fibre thermoplastic (LFT) material under quasi-static tensile and shear loading conditions. In the 0° material direction, a 15% reduction in ultimate tensile strength was observed for 14% lignin content and a 36% reduction was noted for 21% lignin content, compared to 0% lignin content in the flow region. In the charge region, the reductions were 8% and 32.5% for the respective lignin contents.

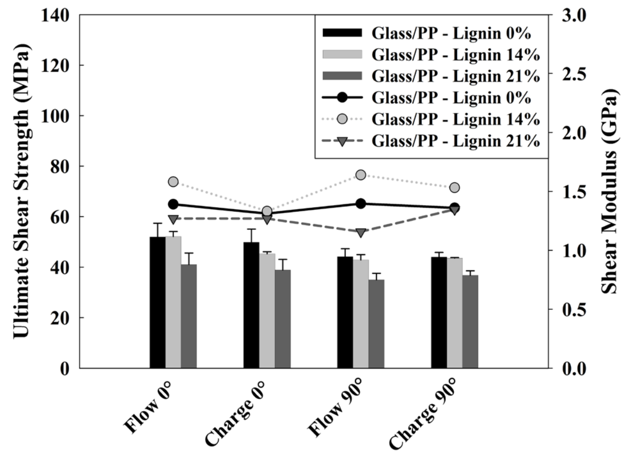

Figure 8. Ultimate shear strength and shear modulus for all material configurations.

The anisotropic nature of the LFT material is evident from the findings, as the tensile strength in the 90° material direction decreased by 76% for 0% lignin and by 55% for 14% lignin, relative to the 0° material direction in the flow region. Similar patterns were observed for Young's modulus, which also exhibited a considerably lower value in the 90° material direction.

For the 21% lignin content, there was a significant degradation in mechanical properties compared to the 14% lignin content. However, for the 14% lignin content, the reduction in mechanical properties was not as drastic, and in some configurations (specifically the 90° material direction), slight improvements were observed. In shear tests, the differences in mechanical properties were less pronounced for all material configurations.

The Glass/PP LFT material with 14% lignin content demonstrated promising results, showing a less significant reduction in mechanical strength compared to material with 21% lignin content. Replacing 14% of the polypropylene matrix with lignin can help reduce the carbon footprint while maintaining acceptable mechanical properties. Additionally, lignin is known to enhance biodegradability, making it a more suitable filler material than other options like calcium carbonate, which does not offer any ecological benefits.

Acknowledgements

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