



Use of lignin as a compatibiliser in hemp/epoxy composites

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ABSTRACT

This study was designed to ascertain if the addition of lignin to hemp-epoxy composites was beneficial to their mechanical properties. Composites were made using a VARTM method with a two-part epoxy resin and a non-woven hemp fibre mat. Lignin was added to the resin before infusion at concentrations varying between 0 and 10% w/w. Samples were then tested according to the relevant ISO standards. There was an increase in impact properties of the fabricated composites with the energy absorbed by the composite containing 5% w/w lignin being 145% higher than the composite with no lignin added. Both flexural and tensile modulus showed an increase when lignin was added up to 2.5% w/w, although there was a drop in both when the lignin was increased to 5% w/w, attributed to poor mixing and infusion due to the increased viscosity of the resin. In all cases, the addition of lignin increased the structural properties of the composites to some degree when compared with composites with no additional lignin.

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1. Introduction

Fibre-reinforced composites (FRCs) are ubiquitous in today's society, in areas such as construction of components for boats, cars and aeroplanes, as well as sports equipment such as tennis racquets and golf clubs. Their benefits include high specific modulus and high specific strength, making them ideal for applications requiring good material properties with low weight. They are made from a combination of a matrix, which can be either a thermoplastic or thermoset polymer, with reinforcements such as glass, carbon, aramid or natural fibres. There has been a wide range of work looking at the properties of these types of composites, either by changing the type of matrix or the type of fibre [1–6].

Two common types of matrix are ester-based (typically polyester and vinyl ester) and epoxy-based resins. The former are normally used in low-performance applications such as bathtubs, shower trays and piping. They do not adhere well to the surface of carbon fibres and are therefore tend to be used in conjunction with glass fibres. For more high-performance composites, epoxy resin is used. When reinforced with boron, carbon and glass fibres its strength is comparable with titanium, steel and aluminium alloys, while showing a significant reduction in weight [7]. Epoxies are commonly used on commercial aircraft and in the manufacture of sporting equipment.

The majority of energy production for industry comes from non-renewable resources, increasing concerns over the volume of fossil fuels used for energy generation. The use of natural materials

which require little energy is therefore desirable. Plant fibres grow naturally, and therefore require minimal man-made energy input. Natural fibres suitable for reinforcement of polymer materials generally contain large amounts of ligno-cellulosic matter. Lignin and cellulose are stringy, tough, wood and plant fibres which help to maintain the structure of plants. Plants high in ligno-cellulosic fibres include hemp, jute (hessian), kenaf, flax, coir, wood and pineapple [8]. Apart from the low process energy required during their manufacture, natural fibres are attractive to create natural fibre-reinforced composites (NFRCs) because of their renewable and sometimes biodegradable characteristics. Even if composting of a particular fibre is not possible it can be burned to recover energy while producing no net increase in carbon dioxide in the atmosphere. This energy recovery process is not possible with glass fibres due to the high temperatures necessary and because of their tendency to cause soiling of the furnace.

However, a disadvantage of natural fibres is their lack of availability as a woven, engineering material. Most natural fibres used currently in the manufacture of biocomposites are made from chop strand mat, and therefore have correspondingly low mechanical properties when compared to woven synthetic fibres such as carbon. However they are generally cheap, widely available and biodegradable. One of the main problems with using natural fibres as reinforcement is the poor interface between the hydrophobic fibres and the hydrophilic resins [9].

When composites were beginning to be developed it was assumed that this interface was able to transmit stresses between the fibre and the matrix perfectly [10], however as photoelastic techniques were developed to allow a visual analysis of stress in a fibre, these theoretical models were found to underestimate the stresses encountered close to the fibre ends [11]. At relatively

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low applied stresses, the shear stresses at the fibre ends of a reinforced composite could exceed the interfacial shear strength, leading to failure by debonding and fibre pull-out.

Fig. 1 shows the deformation that occurs in the area of matrix surrounding a single fibre that is subjected to tensile loading. More recently it has been established that the final mechanical properties of a composite are dependent on the magnitude of the strength of the bond between the fibre and matrix [12–14]. This interface is the limiting factor of fibre-reinforced composite performance as it ultimately defines the amount of load that can be transferred from one fibre to the next by the matrix. A review of some of the research carried out in this field was conducted by Herrera-Franco and Drzal [15]. Work done to model the interfacial bond suggests that there is not a distinct interface between fibre and matrix, but rather an interphase region resulting from the complex chemical interactions between the resin, sizing agents, and the mechanical surface of the fibres [16]. As such, the poor adhesion that exists between the fibres and resins prevents NFRCs from having commercially-useful structural properties.

There are a wide variety of treatments that can be used to improve the fibre–matrix adhesion in composites [4,5,17,18]. The vast majority of these involve some form of chemical processing such as mercerisation with sodium hydroxide solution [19–21] and acetylation with acetic anhydride [9,22–24]. However, it is desirable to reduce the chemical input and associated wastes with the process. There are some alternative treatments e.g. steam explosion [25,26] that avoid some of the chemical input to the composite manufacture; however there is a large associated energy cost with generating the steam required.

An alternative to chemical treatments are the use of natural materials as compatibilisers in the composite structure. Lignin is particularly interesting as it is a waste product from the paper industry [27]. Previous work has shown that lignin can be used as an additive in composite fabrication by RTM; Wool et al. showed that lignin can impart beneficial properties to the structure of a composite by either dissolving the lignin in aqueous sodium hydroxide [28] or chemical modification of the lignin with butyric anhydride to solubilise it in an epoxy resin [29]. However both of these methods still require some additional chemical processing

to the lignin before the composite is manufactured. Lignin has also been utilised in compression moulding techniques to make natural fibre–polypropylene composites [30,31] although this method uses high temperatures which can be potentially damaging to the natural fibre reinforcement and impair the structural properties of the composite.

In this work, it was proposed that even in the solid state, the lignin would improve fibre-to-matrix adherence and structural properties of the resulting composite whilst keeping the number of steps and chemical treatments to a minimum. Hemp fibres were chosen for the reinforcement because of the availability and cost effectiveness of this material in the UK. Epoxy resin was selected as the matrix as this is used in a variety of high performance applications, and therefore has industrially relevant properties.

2. Materials and methods

2.1. General considerations

Kraft lignin was generously supplied by Warwick HRI and dried under vacuum to constant weight before use to remove volatile material. Chopstrand hemp mat was purchased from Hemcore and EP-522 resin and H-522 hardener were purchased from Alchemie.

2.2. Lignin characterisation

Particle Size Analysis was carried out using a Carl Zeiss Axio-Scope A1 upright optical microscope with fully automated stage. A small sample of the lignin powder was placed between glass slides and then mounted in the microscope. Dark field microscopy was used to produce an image with a black background and illuminated particles. The microscope was connected to a PC running Carl Zeiss AxioVision Release 4.8 software and automatic measurement software plug-ins 'MosaicX' and 'Extended Focus' which were used to calculate the effective diameter of each observed particle.

2.3. Composite fabrication

Composite materials were obtained by a VARTM process [32]. An aluminium sheet was prepared beforehand by applying a release film to the surface and coating with silicon release spray to ensure facile removal of the composite after fabrication. A plastic sheet, including a vacuum inlet port and a resin inlet port, was then used to cover the part and sealed with tack tape to make it airtight. The resin mixture was made immediately before use, composed of 100 parts EP-522 and 22 parts H-522. Lignin was then added if desired at the appropriate level (1–10% w/w) and mixed throughout the resin to achieve homogeneity. The mixture was then infused through the fibre mat using vacuum ensuring no air entered the system. Once infusion was complete, both ends were sealed off and the composite was allowed to stand at room temperature for 24 h to cure. The composite part was then post-cured at 120 °C for 2 h and then cut into samples with the desired dimensions for testing.

2.4. Materials testing

Tensile strength and modulus were determined according to EN ISO 527-4: 1997 [33] and flexural strength and modulus were determined according to EN ISO 14125:1998 [34]. Testing was performed on an Instron 5800 universal testing machine equipped with extensometer, with a cross-head speed of 2 mm/min. Charpy impact tests were carried out using a Ray-Ran Advanced Universal Pendulum Impact System following EN ISO 179-2:1999 [35].

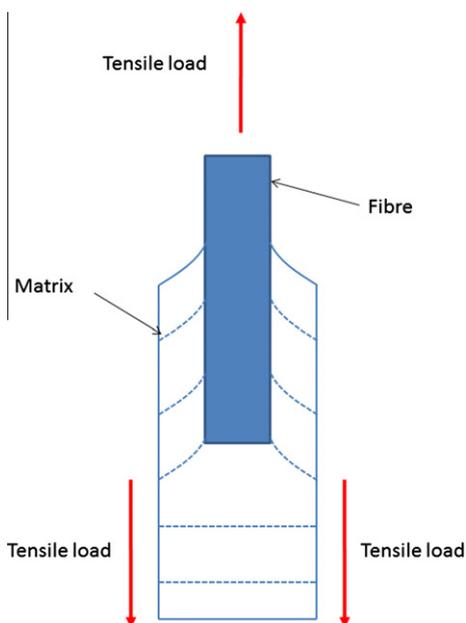


Fig. 1. Effect of application of tensile loading on the matrix surrounding a single fibre.

Table 1
Fibre content of composite samples.

Lignin content (% w/w)	Fibre content (% by mass)
0.0	20.68
1.0	21.82
2.5	21.93
5.0	22.46

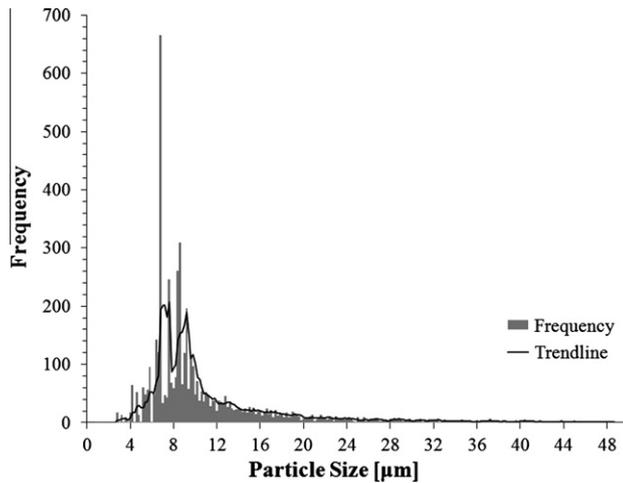


Fig. 2. Distribution of particle size analysis of lignin added to composites.

Results were normalized for sample size and ambient temperature. A hammer with a mass of 1.198 kg, a velocity of 2.9 m s^{-1} and impact energy of 5 J was used for all materials. In all cases, ten samples were used for each test and the results averaged.

2.5. Optical microscopy

In order to investigate the presence of porosity, optical microscopy was used to evaluate images of the structure of the composites. Samples of each composite were prepared by first mounting in Buehler Epcolor red-tinted epoxy resin to give a contrast with the natural fibres. These samples were then prepared by grinding and polishing using a successive grades of P320, P600 and P1200 Silicon Carbide paper were used, followed by final polishing using

0.3 micron Alumina paste. Images of the mounted samples were captured using a Carl Zeiss AxioScope A1 upright optical microscope with fully automated stage. The microscope was connected to a PC running Carl Zeiss AxioVision Release 4.8 software. Zeiss Image Analysis software was used to estimate porosity of the polished samples by calculating the percentage of the image taken up by the red Epcolor resin used to mount the samples.

2.6. Scanning electron microscopy

SEM images of the fracture surface of the tensile test samples were taken and the surface was gold coated by sputtering for 120 s using a current of 25 mA and an approximate coating thickness of 30 nm. The SEM used was a Zeiss Sigma FXM microscope with a field emission gun as the electron source. The acceleration voltage was 15 kV.

3. Results and discussion

3.1. Resin infusion

Hemp–epoxy composites were prepared where the resin had a range of lignin contents (0, 1, 2.5 & 5% w/w). Moisture was removed from the lignin before use by drying to constant weight; using ‘wet’ lignin caused an uncontrollable exotherm in the setting stage and prevented successful infusion. An attempt at a composite with the resin containing 10% w/w lignin was also made but was unsuccessful; addition of lignin to the resin increases the viscosity and at 10% w/w the resin becomes too viscous to infuse in a VARTM process. The viscosity of the standard resin is 700 MPa·s at 25 °C, however this was observed to increase with increasing addition of lignin.

Fibre content (by mass) of all samples was between 20.5% and 22.5%; exact values for each lignin content are shown in Table 1. Previous research has shown that natural fibre reinforced composites have a lower fibre volume fraction compared with synthetic composites manufactured using the same manufacturing method due to difficulties with wet-out and resin infusion caused by the surface roughness and irregularity of the fibres [6].

3.2. Particle size analysis

The frequency distribution of the size of lignin particles used is shown in Fig. 2. Particle sizes of the sample tested ranged from

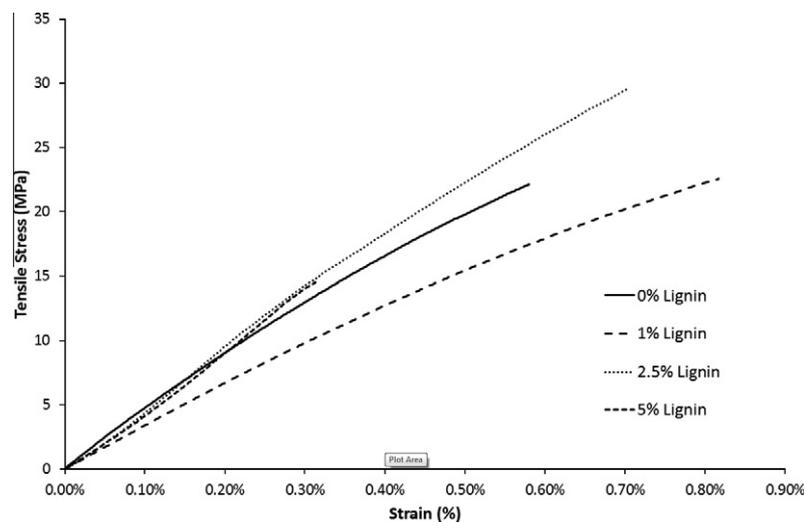


Fig. 3. Tensile stress/strain curves for composites.

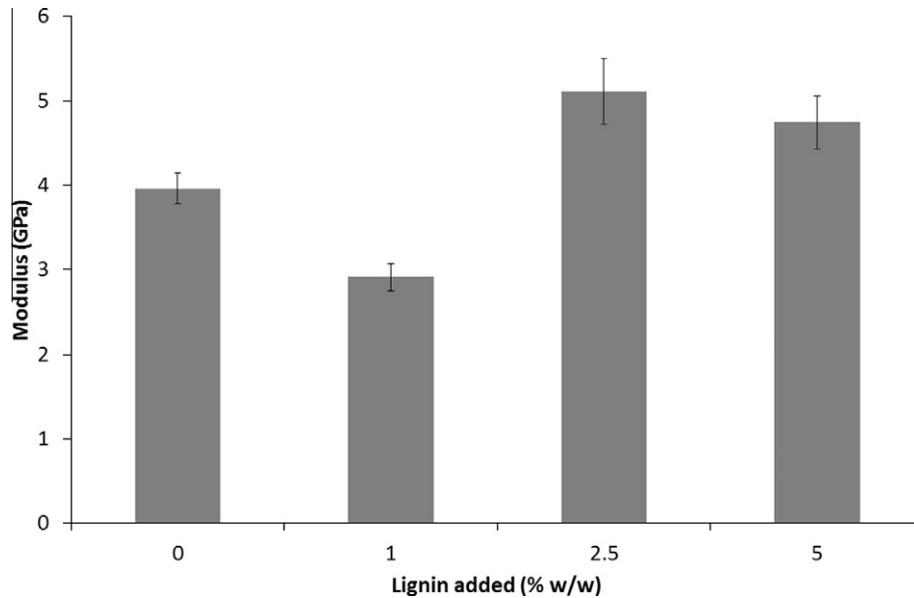


Fig. 4. Tensile modulus vs. lignin additive content for composites.

Table 2

Tensile modulus and UTS for composites.

Lignin content (% w/w)	Modulus (GPa)	UTS (MPa)
0.0	3.96 ± 0.18	22.63 ± 2.11
1.0	2.91 ± 0.16	20.57 ± 1.15
2.5	5.11 ± 0.39	31.15 ± 2.71
5.0	4.74 ± 0.31	23.37 ± 2.24

1.6 micron up to 50 micron, with 65% of the particles having a diameter of between 5 and 10 microns. The perceived wisdom is that the smaller particles are more beneficial to the process as they are able to more effectively penetrate the non-woven hemp fibre mat. Further processing could be used to refine the particle size, for example micronizing or filtration, however the added complexity and energy usage of these techniques was considered detrimental to the overall environmental impact of these composites.

3.3. Tensile testing

The results for the tensile testing are shown in Figs. 3 and 4 and Table 2. In all cases, it would appear that 2.5% w/w lignin is the optimum amount for improving the properties of the final part. The Young's modulus increased from 3.96 GPa at 0% w/w to 5.11 GPa at 2.5% w/w and the Ultimate Tensile Strength (UTS) increased from 22.6 MPa to 31.1 MPa for the same addition of lignin. A further increase to 5% w/w lignin content caused a 7.4% reduction in Young's modulus and a 25.0% reduction in UTS, most likely caused by the large volume of lignin particles preventing complete wetting out of the fibre reinforcement [28]. This would also explain the brittle nature of the composite, given that the strain at failure is on average 27% higher for composites containing 2.5% w/w lignin than 5% w/w lignin.

There are some interesting changes when lignin is added at a 1% w/w level (Fig. 4). The ductility of the composite is increased compared with both 0% w/w and 2.5% w/w lignin content. However, the tensile strength is reduced indicating there is a plasticising

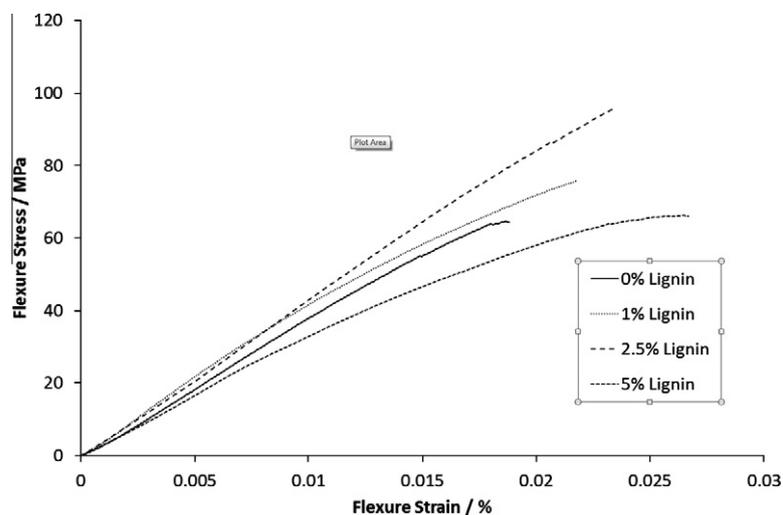


Fig. 5. Flexural stress/strain curves for composites.

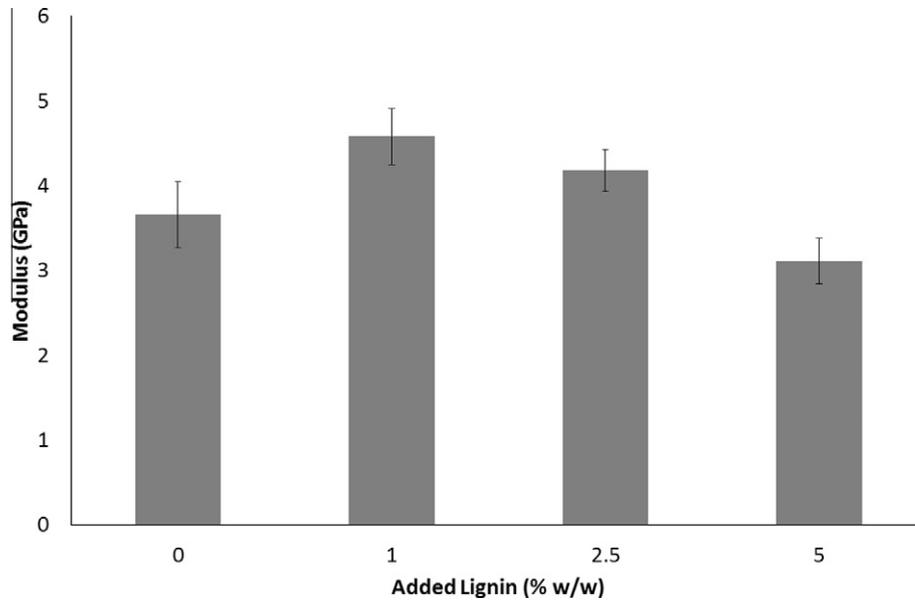


Fig. 6. Flexural modulus vs. lignin additive content for composites.

3.4. Flexural testing

An increase in strength was also observed when the composites were tested for their flexural stress properties (Figs. 5 and 6). The maximum flexure stress was observed at 2.5% w/w with a value of 86.16 MPa, which was a 31.0% increase on the composites made with no lignin content. There is an increase in both strength and ductility for both the 1% w/w and the 2.5% w/w composites, indicating good mixing and infusion. The composites containing 5% w/w lignin have retained the ductility of the composites with smaller amounts of added lignin, but the strength is decreased to below that of the 0% w/w composite; 60.0 MPa compared with 65.7 MPa (Table 3). Poor mixing due to the increased viscosity would also account for this drop in strength producing brittle failure, similar to that exhibited under tensile strain but less critical under shear flexure.

Table 3
Flexural results for composites.

Lignin content (% w/w)	Modulus (GPa)	Max. stress (MPa)
0.0	3.66 ± 0.39	65.77 ± 6.65
1.0	4.58 ± 0.33	78.07 ± 1.99
2.5	4.18 ± 0.25	86.16 ± 5.11
5.0	3.11 ± 0.27	60.00 ± 4.16

effect of the lignin at that level; increased flexibility but reduced strength and stiffness. This can be attributed to a combination of the interfacial effect between the lignin and the hemp fibres and an effect related to the increased viscosity of the resin causing incomplete infusion of the composite part.

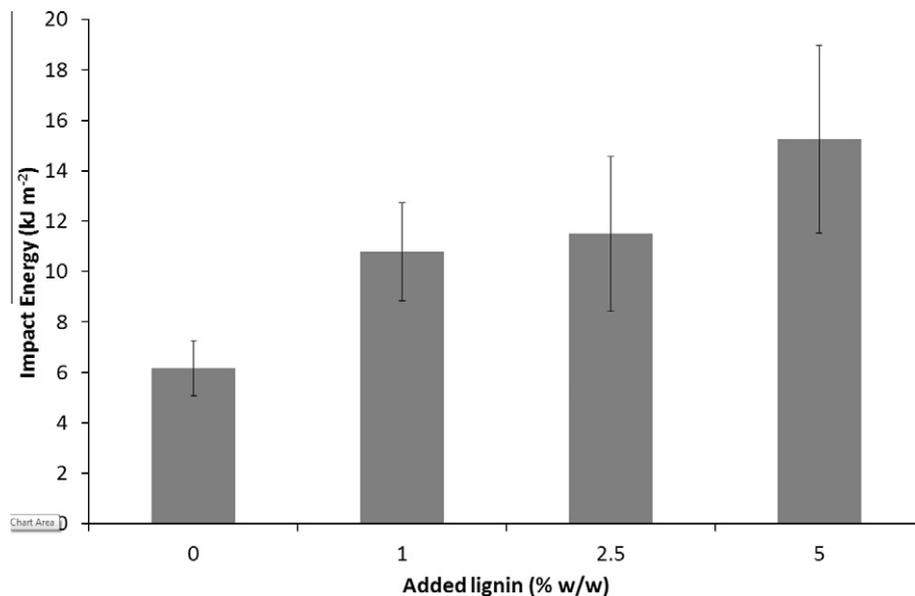


Fig. 7. Impact energy vs. lignin additive content for composites.

The flexural modulus showed a similar pattern although the peak was observed at 1.0% w/w lignin content instead of 2.5% w/w. The flexural modulus of composites with lignin contents of both 1.0% w/w (4.58 GPa) and 2.5% w/w (4.18 GPa) showed an improvement on the composite made with no lignin added (3.66 GPa), again highlighting the beneficial properties of lignin in natural fibre composites. The addition of 5% w/w lignin proved to be damaging to the composite properties again as it resulted in a drop in modulus to 3.11 GPa, a value below that of the composite with no lignin content.

3.5. Impact testing

Comparison of the impact properties of the fabricated composites using the Charpy impact test showed that there is an increase in toughness as more lignin was added to the part (Fig. 7). With no lignin added the Charpy impact strength was 6.17 kJ m^{-2} , which increased to 10.78 kJ m^{-2} when 1.0% w/w lignin was added. Addition of a further 1.5% w/w showed very little improvement giving a value of 11.50 kJ m^{-2} for the impact strength. However, increasing the lignin content to 5.0% w/w showed a significant rise in the impact strength of the composite (15.24 kJ m^{-2}).

3.6. Optical and scanning electron microscopy

Scanning Electron Microscopy was used to analyse the failure surfaces of the hemp/epoxy samples after they were subjected to Charpy Impact testing. Fig. 8 shows a sample with no added lignin which has evidence of significant fibre pull-out and a lack of



Fig. 8. SEM image of hemp/epoxy composite with 0% w/w lignin added.

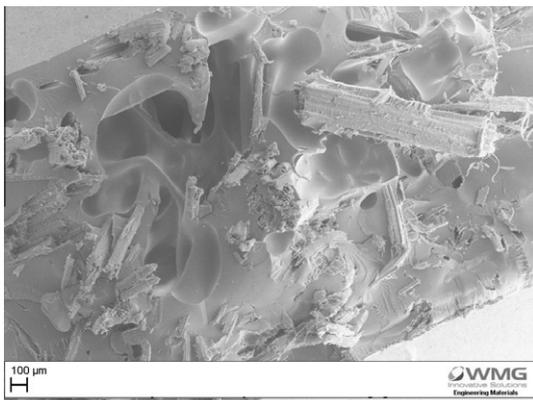


Fig. 9. SEM image of hemp/epoxy composite with 1% w/w lignin added.

interfacial adhesion. Fig. 9 shows the sample with 1% lignin added. This agrees with the reduction in mechanical properties identified after mechanical analysis, and the authors suggest the cause to be the increase in viscosity noted during infusion.

At 2.5% added lignin, fibre pull-out is reduced and there is evidence of partial fibres remaining attached to the matrix (Fig. 10). Adding 5% lignin (Fig. 11) appeared to further increase the interfacial bond strength, as there are instances of matrix failure around fibres and a reduction in instances of fibre pull-out. The reduction in tensile strength compared to 2.5% lignin addition can be attributed to the increase in porosity which has been shown to have an effect on the mechanical properties of composite materials [36,37]. This effect has been previously observed by Thielemans et al. [28], and is a result of large volumes of lignin particles preventing complete wet-out of the fibre reinforcement.

Figs. 12–15 show images captured by dark field optical microscopy. All samples show evidence of porosity signified by the red colour of the resin used to mount the samples. The porosities of each composite are given in Table 4.

Optical microscopy confirmed that the sample with 1% lignin added showed higher porosity than the other samples tested,

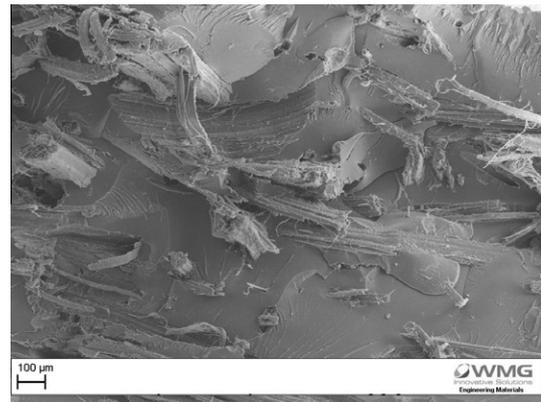


Fig. 10. SEM image of hemp/epoxy composite with 2.5% w/w lignin added.

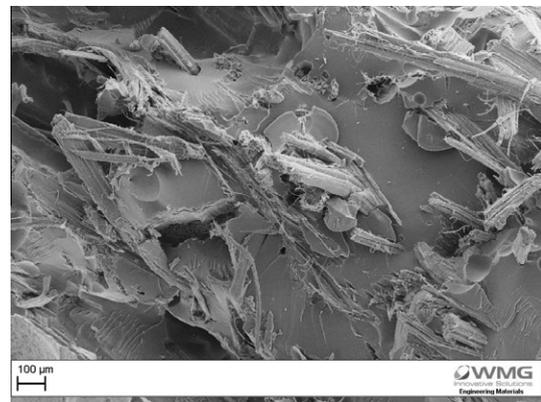


Fig. 11. SEM image of hemp/epoxy composite with 5% w/w lignin added.

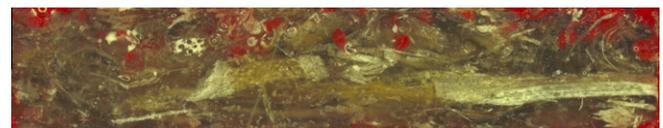


Fig. 12. Dark field optical micrograph of hemp/epoxy composite with 0% w/w lignin.

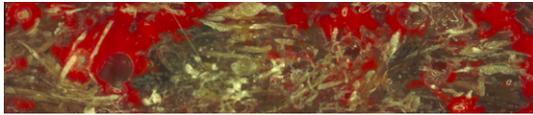


Fig. 13. Dark field optical micrograph of hemp/epoxy composite with 1% w/w lignin.



Fig. 14. Dark field optical micrograph of hemp/epoxy composite with 2.5% w/w lignin.



Fig. 15. Dark field optical micrograph of hemp/epoxy composite with 5% w/w lignin.

Table 4
Porosity of composite samples.

Lignin content (% w/w)	Porosity (%)
0.0	4.2
1.0	12.8
2.5	1.49
5.0	1.73

caused by the increased viscosity of the resin from the addition of lignin (Fig. 15).

4. Conclusions

Composites made from an epoxy resin and natural hemp fibre reinforcement with varying amounts of added kraft lignin were fabricated. The addition of lignin was shown to be beneficial towards improving the impact, tensile and flexural strength, although the latter two also showed a decrease when excessive lignin was added. This is attributed to the lignin particles preventing complete resin infusion across the hemp mat which subsequently reduces the physical properties. This method of adding lignin to composites has advantages over previous methods as it eliminates the need for costly and wasteful chemical processing, either to the natural fibre mat or the lignin itself to solubilise it in the resin matrix.

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