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Use of interleaved films to enhance the properties of all-cellulose composites

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ABSTRACT

All cellulose composites (ACCs) were produced by partially dissolving alternating layers of cotton textile and cellulose film in solvent solution and heating them under an applied pressure. The ionic liquid 1-ethyl-3-methylimidazolium acetate ([C2MIM][OAc]) combined with a co-solvent dimethyl sulfoxide (DMSO) was used in this work, and an iterative approach was employed to identify the combination of [C2MIM][OAc] and DMSO to yield the best balance of composite mechanical properties. It was revealed that the addition of the interleaved film yields a higher Young's modulus and significantly higher interlayer peel strength compared to processing without the film. With an optimised [C2MIM][OAc] and DMSO combination, Young's modulus was improved from 2.2 ± 0.2 GPa to 4.2 ± 0.2 GPa and peel strength reached as high as 917 ± 2 N/m. In addition, there was improved uniformity between longitudinal and transverse mechanical properties, which we attribute to an overall increase in interlayer adhesion.

1. Introduction

ACCs have attracted significant attention within the research community in recent years as sustainable alternatives to traditional composites [1–3]. In this context, ACCs are composites where both fibre and reinforcement are comprised entirely of cellulose, a naturally occurring biopolymer with inherently good mechanical properties [4,5]. Concerns have continued to rise regarding the environmental impact of manufacturing industries, and so there has been a growing eagerness to drive innovation towards developing sustainable materials that can better support a circular system.

Composites combine the characteristics of two or more different materials to achieve improved, or enhanced properties [6]. In general terms a composite comprises a continuous phase known as the matrix, which surrounds a dispersed phase known as the reinforcement. Crucially, there must be excellent adhesion between the matrix and reinforcement phases for the composite to achieve an excellent balance of mechanical properties. Having chemically identical components gives ACCs an advantage over traditional fibre reinforced polymer composites (FRPCs) where recycling mixed material components can be challenging [3].

It is well understood that cellulose does not melt [4,7–9], and so to create a continuous matrix that can infuse into and then bind to a reinforcing fibre assembly, requires dissolution and subsequent coagulation after solvent removal. There are two established ways to produce ACCs; the first method initially conceived by Nishino et al. [4] involves the dissolution of cellulosic fibres in solvent to form a solution into which further cellulosic fibres are immersed. This method was later adapted by Gindl et al. [5] to a selective dissolution process of a single component, the fibre assembly itself, where the outer layer of cellulosic fibres is partially dissolved to form the matrix phase after coagulation, and the inner core remains undissolved which serves as reinforcement [5,10,11]. The advantage of such a single component process is that the matrix is created around each reinforcing element, removing any issues with impregnation, which can often be a significant challenge when using a dense woven textile layer.

The developments in ACC research have been summarised within the scientific community [1,2,12], highlighting the gaps in present understanding as well as areas warranting further development; one particularly significant area noted was the limited thickness of ACCs. Owing to the process of dissolution in solvent and subsequent solvent removal, most studies into preparing ACCs result in thicknesses <1 mm, and to broaden the range of potential applications of these composites it would be necessary to produce thicker structures [1,13].

The use of textiles as reinforcements for ACCs has been explored as a fresh approach to achieve thicker ACCs [3,6,14,15], where layers of

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textile are partially dissolved in solvent [6,14,15], or added to a pre-dissolved cellulose solution [3]. Achieving adequate adhesion between individual layers can be a concern for all composite laminates, however, as poor interfacial bonding between chemically incompatible components can lead to inefficient stress transfer [16] and delamination, a failure mechanism to which layered materials are particularly susceptible [1,14,17]. This has been a potential difficulty for traditional heterogeneous composites, and indeed natural fibre reinforced composites (NFRCs), where the opposing polarities of a thermoplastic or thermosetting polymer matrix and natural fibres can often create a weak interfacial bond [3].

Although the homogenous nature of ACCs can help to improve interfacial adhesion between fibre and matrix phases, it is still a challenge to achieve sufficient bonding between layers of textile and surrounding matrix, particularly when partially dissolving the fibres within the textile. A reason for this can be suggested by looking at how bonding is achieved when thermoplastic polymer fibres are selectively melted under the application of heat through a process termed hot compaction. Here the outer skin of each polymeric fibre is selectively melted, which on cooling forms the matrix phase of the single polymer composite [18]. The fraction of melted polymer, however, is not always sufficient to adequately bond layers of woven polymer filaments, owing to the pockets of space that exist between the individual textile layers, due to the textile surface roughness. More material is therefore needed to produce a matrix that can achieve adequate bonding, but only in the intra-textile layer, as in the absence of this, delamination can occur [14]. Our previous research on thermoplastic single polymer composites [18] showed that one way to achieve this balance is to incorporate an interlayer film, of the same chemical composition, which significantly increases the interlayer strength without requiring significant loss of the reinforcing fibres. A similar situation can be attributed to the current work on the partial dissolution of cellulosic fibres within a textile, where a layer of dissolved material around each fibre surface is produced, forming the matrix to aid fibre to fibre bonding. The amount of cellulose that can be dissolved is highly influential in determining the matrix fraction of the resulting composite, as remarked in previous studies into ACC production [6].

To increase matrix fraction and enhance interlaminar bonding, several approaches can be taken. One approach involves consideration of the solvent, which can play an important role in supporting many discussed mechanisms involved in cellulose dissolution, for example inter- and intra- molecular bond breaking [19,20], polymer chain disentanglement [21], and hydrophobic interactions[22,23]. There has been much attention given recently into the use of ionic liquids (ILs) [8,14,15,24,25]. ILs are a class of solvents with favourable attributes such as low vapour pressure, high thermostability, and the potential to be recycled [26]. Another approach forming the backbone of this work comes from our previous thermoplastic single polymer composite research as described above. This involves incorporating additional matrix in the interlayer regions from a second cellulosic source (in this case thin cellulosic film), akin to the two-step method described above. A version of this was demonstrated in the work of Spörl et al. [3], where a layer of pre-dissolved cellulose solution was added between layers of cotton textile.

In this present work, the addition of an interleaved cellulose film between layers of woven cellulosic textile is investigated in a modified partial dissolution process. By placing matrix material in-between textile layers, it is suggested that matrix production can be targeted to where it is needed the most; the interlayer regions which can be a weak point in multiple layer composites. This could improve interlaminar adhesion and limit overall fibre dissolution, allowing the mechanical properties of the textile reinforcement to be utilised more efficiently. Initially, the effect of the interleaf film between two layers of cotton textile was studied by comparing ACCs produced under similar processing conditions, using different amounts of solvent solution, and compared to samples made without film. It is important to acknowledge the different cellulosic sources used in this work, native cellulose in the cotton and regenerated cellulose film. It is known that the mode of dissolution can depend strongly on the ability of the solvent to disrupt the long-range order of cellulose chains [27], and the higher degree of polymerisation (DP) of native cellulose when compared to regenerated cellulose can lead to reduced polymer mobility in solution and a slower dissolution rate. Although these factors could influence the readiness of different sources of cellulose to dissolve, in this work it is suggested that the amount of solvent has a more prominent influence on the overall cellulose dissolution owing to the high temperature conditions. Previous work within our research group has shown that the dissolution of cotton obeys time–temperature superposition [28]. In the present study, dissolution time and temperature are fixed parameters of 10 min and 100 °C respectively. By processing at an elevated temperature, it is suggested that as dissolution occurs so quickly, the dissolution is instead controlled by the amount of solvent used, rather than time and temperature. The concentration of dissolved cellulose in the surrounding solution is therefore higher than that of the previous study, where the yarns were in excess solvent [28], and subsequent solution viscosity will be higher, affecting the kinetics. Within this duration of 10 min, a higher fraction of the film may be dissolved than the textile owing to it being considerably thinner (0.023 mm compared to 0. 229 mm respectively) which leads to a higher surface area to volume fraction. The cotton may appear to dissolve slower because it is thicker, although the absolute amount dissolved may be similar.

Subsequent iterations were then performed with a view to establishing whether the addition of a co-solvent would be beneficial for this process. It is widely acknowledged that ILs possess a high viscosity, increasing further when combined with a polymer such as cellulose [29]. DMSO is a colourless, aprotic solvent of low toxicity, formed as a by-product from the production of paper from wood pulp [30]. It has been identified as a suitable co-solvent to mix with IL's when dissolving cellulose as it can reduce the viscosity of the IL without compromising its ability to form solvent–solute bonds with cellulose [29,31]. The solvation power of imidazolium based ILs can also be improved with the addition of DMSO and there are various possible explanations cited as to why this might be the case. On one hand it is believed that the ability of DMSO to reduce viscosity can increase IL solvation power by assisting mass transport [32], but the ability of DMSO to modify the diffusion coefficients of the cation and anion in these ILs has also been considered [31]. With this existing knowledge in mind, it seems worthwhile to explore the effect of DMSO on the resulting ACCs, and whether its presence is beneficial when applied to hot compaction, where pressure and elevated temperatures are involved. While the addition of DMSO might favourably adjust the diffusive mechanisms of the ions, the reduction in viscosity may indeed cause solvent to be lost under pressure, reducing the presence of dissolved cellulose and subsequent matrix. ILs are, however, typically expensive to synthesise [33], and so to use a dilution in DMSO would be of additional benefit in lowering processing costs associated with their use. It might therefore be suggested that to a point, adding DMSO is advantageous, and confirming an optimum concentration for this process is highly valuable.

In the final part of the study, the effect of the interleaf film on multi-axial properties was explored. It is generally understood that fibre orientation plays a crucial part in optimising the tensile properties of ACCs [34], and unidirectional ACCs, where the fibres are aligned in one direction, can exhibit excellent tensile properties in the longitudinal direction of the fibres. There is, however, a drawback to unidirectional aligned reinforcement fibres where properties are significantly weaker in the transverse direction, due to fibre alignment on just one axis. The orientation of fibres on two axes in the case of a woven fabric can provide more balanced properties in the fabric plane [35]. This would widen the potential applications of ACCs. The use of woven textiles as a composite preform provides yarn orientation in two directions owing to the interlaced ends (warp yarns) and picks (weft yarns) aligning perpendicular to each other, thus providing a (0°, 90°) orientation. It can be said, therefore, that the warp yarns contribute to the longitudinal properties of a woven textile, and the weft yarns contribute to the transverse properties [36]. In some woven structures, particularly non-technical textiles, the warp yarns typically have a higher tensile strength than weft yarns [37] and this relates to the degree of crimp present in the yarns after weaving. During the weaving process, warp yarns are held under tension and weft yarns are passed under and over at 90°. The interlacing of the yarns leads to a degree of waviness known as crimp [38] and is generally more pronounced in the weft yarns. When applying load to a textile, the initial load is used in straightening out the yarns, leading to reduced tensile properties in the direction of the weft yarns [39]. The strength of the textile will be transferred to the composite it reinforces, and so strength may not be equal in these directions. Commercially obtained fabrics such as the cotton used in this study are more likely to have an imbalance of properties as described above, whereas technical textiles made specifically for composites typically possess more balanced properties in the direction of the warp and weft yarns. It is proposed that by using an interleaved film, the additional matrix produced will improve the efficiency of the stress transfer between longitudinal and transverse orientated yarns by improving the overall level of adhesion throughout the composite structure.

2. Materials and methods

2.1. Materials

Commercially available bleached 100% cotton with a plain weave (obtained from Oh Sew Crafty, Kidderminster, UK) was used as the cellulosic precursor fabric in this study. The fabric had an areal density of 136 g/m^2 and a thread count of 120 in both directions. Natureflex 23NP cellulose film supplied by The Futamura Group, was used as the cellulosic interleaf layer and has a thickness of 23 µm. The films were supplied containing small amounts of additives that were not removed prior to processing; these additives did not appear to disturb the dissolution process. [C2MIM][OAc] with purity of ≥95% was purchased from ProIonic and dimethyl sulfoxide with a purity of ≥99.9%, was purchased from Fisher Scientific.

2.2. Composite processing

ACCs were prepared by introducing a solution of [C2MIM][OAc] and DMSO to a layered stack of cotton fabric and Natureflex film, and heating under pressure in a laboratory heat press. ACC samples were made using two layers of textile with a single layer of interleaf film in between. 10 cm × 10 cm layers of cotton textile were cut and weighed to obtain an overall mass of cellulose to be processed, and the mass of solvent solution to apply was then determined. Solvent to cellulose (S/C) weight ratios of 1:1, 2:1 and 3:1 were used to explore the optimum amount of solvent required to allow the best distribution to the textile layers. The S/C weight ratios were calculated based on the weight of the cotton textile only. Initially a solvent solution comprising 80% by weight [C2MIM][OAc] and 20% DMSO was used, and in a subsequent iteration, the percentage [C2MIM][OAc] in DMSO was varied from 100% (neat [C2MIM][OAc]) down to 20% to establish whether the initial 80% [C2MIM][OAc] concentration was the best choice. The solvent mixture was applied to the first textile layer using a brush, after which the layer of cellulose film was added and pressed down. The second textile layer was then added and the remaining solvent applied. Each textile layer was treated with equal amounts of solvent to ensure even distribution of the solution. The layered stack was placed in a laboratory heat press at 100 °C with an applied pressure of 2 MPa for 10 min. After this partial dissolution process, the samples were placed in a coagulation bath of distilled water at room temperature and left for 20 h to remove the solvent. After coagulation and solvent removal, the stack was dried in the heat press for 1 h at 125 °C and 2 MPa. Fig. 1. shows a schematic of the process stages.

2.3. Cloth orientation during sample preparation.

For the first set of iterations, samples were made with both cotton textile layers stacked at 0° with respect to the warp yarns, giving a stacking sequence of (0,0). Once the appropriate solvent configuration had been established, additional ACC samples were made with film for comparison to explore how the cloth orientation within the stack influenced properties. A two-layer sample was prepared with one layer of cloth orientated 0° relative to the warp yarn direction, and the second layer orientated 0° relative to the weft yarn direction (0,90). A fully symmetric sample was prepared using 4 layers of cloth, with the warp yarns aligned 0°, 90°, 90°, 0° from bottom to top (0,90,90,0). The different stacking arrangements used throughout this study are summarised in Fig. 2.

2.4. Mechanical testing

Tensile strength (ASTM D1846), Young's modulus (ASTM D1846) and peel load (ASTM D1846) were evaluated using an Instron 5584

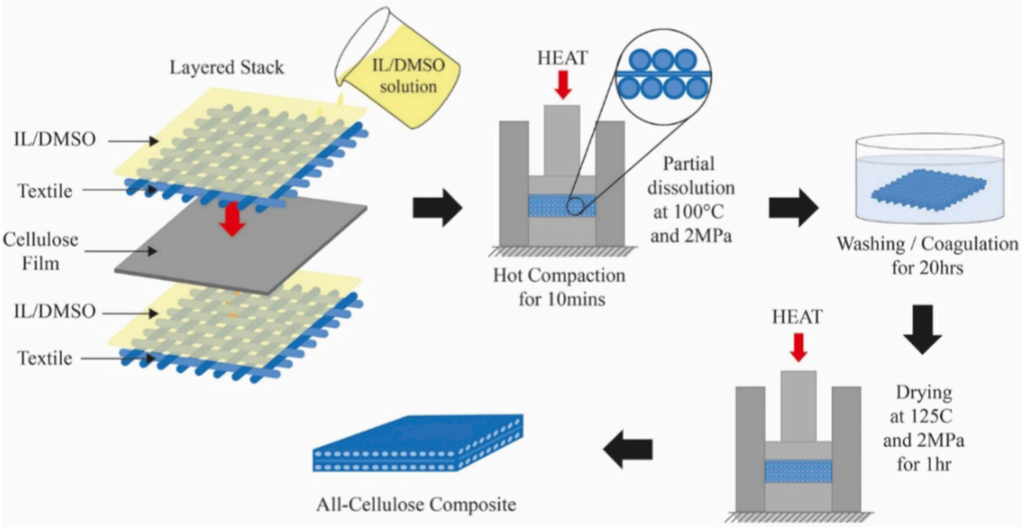

Fig. 1. Schematic outlining the various stages in the manufacture of ACC samples using cotton cloth and cellulosic film layers. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

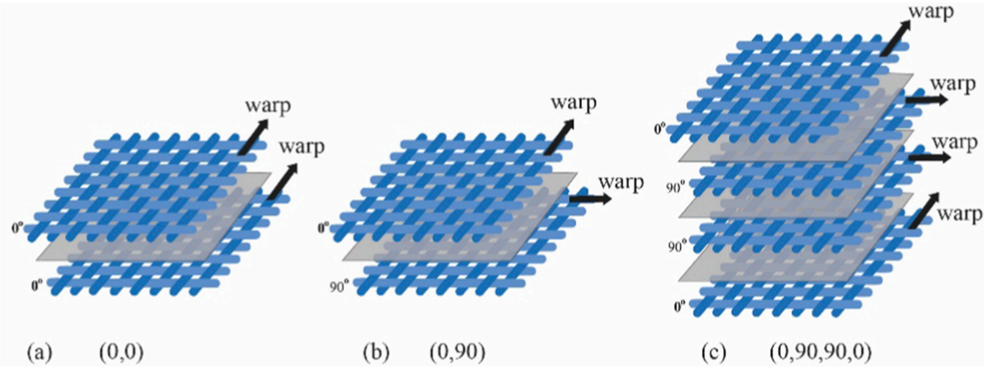

Fig. 2. Stacking sequences of cloth textile layers used in the study. (a) shows a 0,0 stacking arrangement where the warp yarns of both layers are aligned in the same direction, (b) shows a 0,90 stacking arrangement where the first and second layers are aligned at 0° and 90° respectively, and (c) shows a 0,90,90,90 symmetric stacking arrangement where the upper and lower textile layers are aligned at 0°, and the middle layers are aligned at 90°. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

universal tensile tester. To determine Young's modulus and tensile strength, specimens of width 5 mm and a gauge length of 30 mm were tested using a crosshead speed of 10 mm/min. Five specimens were cut from each sample where an average value and corresponding standard error could be calculated. For the peel tests, specimens had a width of 10 mm and a length of 80 mm and were tested at a speed of 80 mm/min. Prior to the dissolution process, a thin layer of foil was placed in between the two textile layers at the edge to provide an unbonded region from which to start the peel test. ACCs were initially tested in the direction of the warp yarns (longitudinal), with subsequent tests carried out in the direction of the weft yarns (transverse), and 45° to the warp yarns.

2.5. Materials characterisation

2.5.1. Optical microscopy

Preliminary surface images of the prepared ACC samples were obtained using a Swift 350B optical microscope in transmission mode. Reflection mode was used to explore the cross sections of the ACCs prepared with and without film. Multiple images of each sample were collected to ensure the surface characteristics were consistent across the sample size. ImageJ was used to obtain measurements of the ACC thickness and cross-sectional area (CSA) of the fibres present in the samples. Six measurements of thickness and CSA were taken to calculate an average value and corresponding standard error.

2.5.2. ACC density calculations

ACC specimens were cut and measured using an RS PRO digital caliper to obtain accurate width and length measurements. Using thickness measurements as described above, specimen volume was calculated before weighing to obtain density measurements of the ACCs. From previous literature, the absolute density of plant fibres is reported to be between 1.4 and 1.5 g/cc [40], the density of cellulose II is estimated as 1.530 g/cc [41], and bulk density of amorphous cellulose has been reported to be 1.48–1.5 g/cc [42]. It was therefore proposed that a value of 1.5 g/cc would be used for the density of the cellulose components within the ACC, for comparison purposes.

2.6. Viscosity measurements

Viscosity measurements of the of [C2MIM][OAc]/DMSO mixtures were taken using an Anton Paar MCR 302 Rheometer at 25 °C and 100 °C. Two measurements were taken to obtain an average and corresponding standard error.

2.7. Estimation of cellulose content of processed ACCs

There are some challenges faced when estimating fibre volume fraction of ACCs produced via partial dissolution methods. Unlike traditional fibre reinforced composites, the volume of fibres used to prepare ACCs will not be the same after processing as a certain amount of fibre will have dissolved to become the matrix. Additionally with the dissolution process being carried out under pressure, there will also be some dissolved cellulose pushed out from the mould plates, and this is known as flashing. Estimating the fibre volume fraction of the ACCs is therefore more complex, and so quantitative analysis of crystalline structure using X-Ray Diffraction (XRD) was employed. Before dissolution, the majority of cellulose I and cellulose II will be present in the cotton cloth and interleaf film respectively, together with an amorphous fraction. After partial dissolution, the dissolved fractions of cotton and film will be converted to amorphous cellulose and cellulose II. Using this knowledge, XRD could then be employed to characterize the change in the crystalline structure of ACC samples made with and without interleaved film and provide estimations of the cellulose I, cellulose II and amorphous cellulose fractions present in the ACCs. Using a Bruker D8 Diffractometer with monochromated Cu Kα_radiation, scans were run in Gonio (Bragg-Brentano) mode to get a diffraction pattern. 2θ line scans were run from 5° to 70° with a step change of 0.013°. Samples were scanned in their solid form to perform the analysis on the most representative version of the ACC structure, and to avoid any potential degradation or crystal modification that might arise through grinding. To ensure a homogenous representation of the sample was seen, the sample stage was spinning at 2 revolutions per second. Estimated fractions of cellulose I, cellulose II and the amorphous component were calculated using a deconvolution method [28]. Specimens were cut from three different locations within the sample to allow average values and a corresponding standard error to be calculated. Cellulose I is known to show peaks at 14.8°, 16.3°, 20.6° and 22.4° and cellulose II has peaks at 12.4°, 20.2° and 21.8° [28,43]. Amorphous cellulose is also known to have a broad peak around 18° [43,44].

3. Results and discussion

3.1. Influence of interleaf film

The longitudinal (warp direction) mechanical properties of ACCs produced with and without an interleaf film using various S/C weight ratios are presented in Fig. 3. Error bars shown in Fig. 3 (a) and Fig. 3 (b) reflect the standard error calculated across the five tested specimens, and errors bars in Fig. 3 (c) reflect the variation of the measured peel strength along the sample. Fig. 4 shows surface and cross-sectional images of all prepared ACCs including the original raw cotton textile for comparison. Values for ACC thickness, density, and yarn CSA are also presented in Table 1.

During preparation it was found that when using an equal weight of solvent to cellulose (1:1), there was scarcely enough solvent to distribute to the textile layers. Consequently, there is no significant change in properties between the samples made with film and without, and the larger errors present for these samples indicate the variation in properties across the sample due to inconsistent solvent distribution. The woven structure of the cotton cloth allows for rapid absorption of the

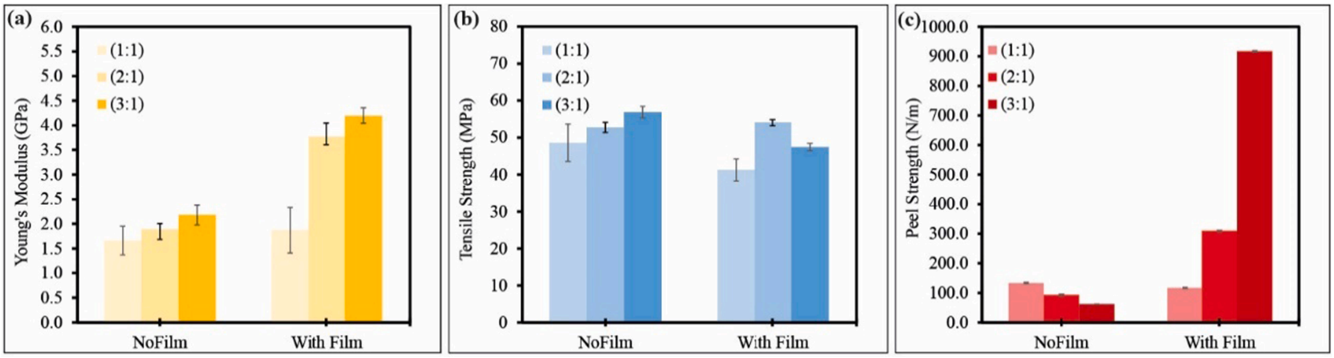

Fig. 3. Young's modulus (a), tensile strength (b), and peel strength (c) of all-cellulose composites prepared with interleaf film and without interleaf film using various S/C weight ratios. Samples were processed for 10 min using an 80/20 ratio of [C2MIM][OAc]. Mechanical properties were tested in the longitudinal direction. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

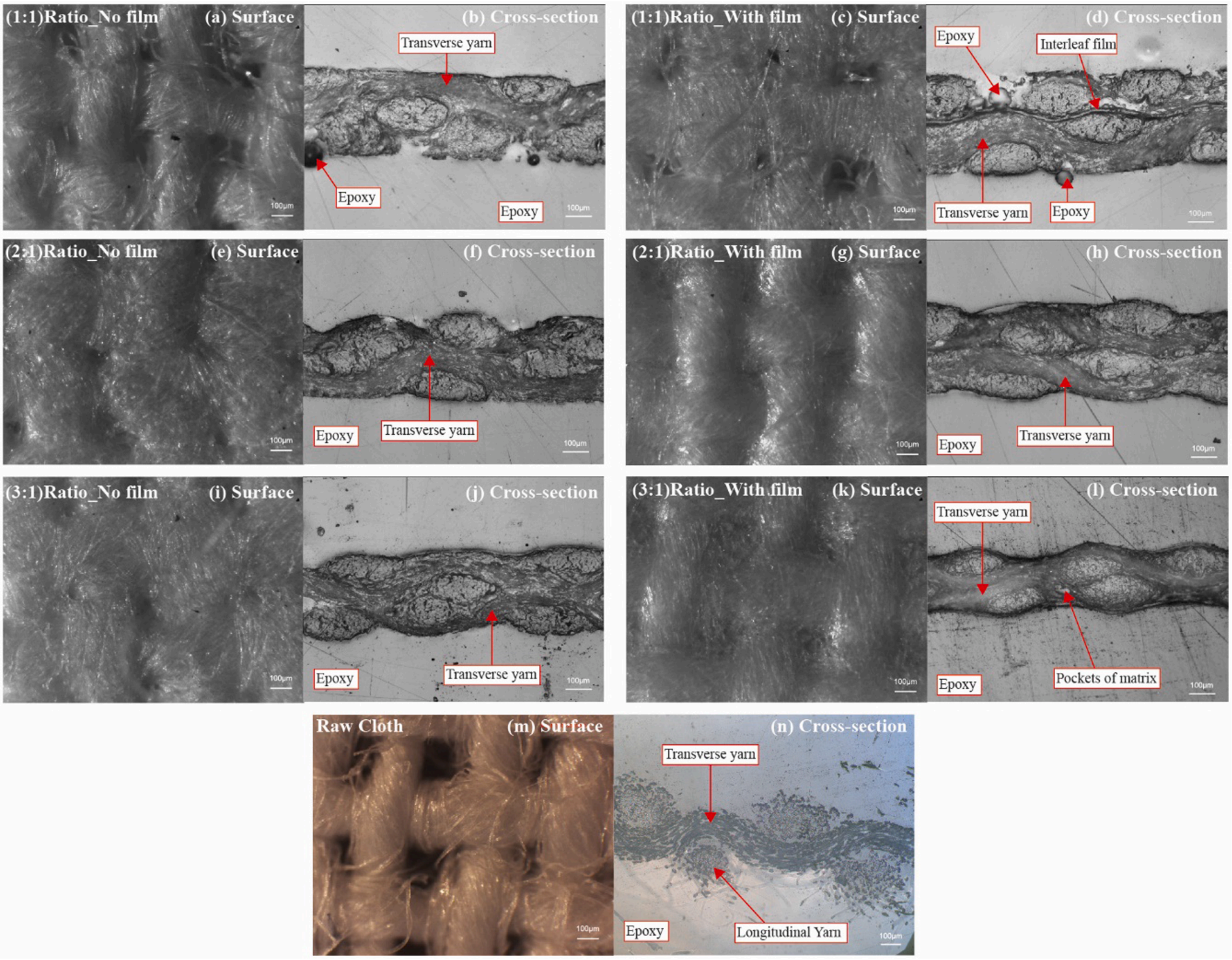

Fig. 4. Optical microscopy images of ACCs prepared without film (a, b, e, f, i, j) and with film (c, d, g, h, k, l) using various S/C weight ratios of 1:1, 2:1 and 3:1. Raw cotton (m) and (n) is also shown for comparison. Surface images and cross-sections are displayed for each sample. The darker regions within the sample cross-section indicate the presence of void space, as indicated by the red arrows. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

solvent solution into the pockets of space between interlinking yarns, making even distribution of the solvent difficult to achieve when using less solvent. This suggests that there is insufficient solvent to partially dissolve enough fibre and film to produce the required matrix for a composite structure, and this is further evidenced in Fig. 4(b) where the inconsistency in appearance can be seen across the ACC cross-section. Whilst this may result in a higher fibre volume fraction, the lack of useful matrix leads to inefficient stress transfer to the reinforcing component and decreased mechanical properties. The interleaf film can also still be seen clearly as shown in Fig. 4(d), and there is little

Table 1
Calculated values of thickness (mm), density (g/cc), and yarn CSA (mm^2) for ACCs prepared with and without interleaf film using various S/C weight ratios. Thickness and density of the unprocessed raw materials are also presented.

S/C Ratio	Preparation	Thickness (mm)	Density (g/cc)	Yarn CSA (mm^2)
(1:1)	No film	0.30 ± 0.01	0.96 ± 0.02	0.025 ± 0.002
	With film	0.36 ± 0.01	0.87 ± 0.02	0.0243 ± 0.0002
(2:1)	No film	0.29 ± 0.01	1.04 ± 0.01	0.028 ± 0.001
	With film	0.34 ± 0.01	0.92 ± 0.04	0.027 ± 0.001
(3:1)	No film	0.31 ± 0.01	0.91 ± 0.016	0.029 ± 0.004
	With film	0.22 ± 0.005	1.31 ± 0.02	0.015 ± 0.004

Raw materials	Thickness (mm)	Density (g/cc)
Raw cotton textile	0.229	1.5
NatureFlex 23NP film	0.023	1.5

difference between the ACC surfaces from using the film or not except for a flattening of the yarns under compaction. Film thickness in this sample was measured to be 0.0087 ± 0.0003 mm, suggesting that although some dissolution of the film has occurred (over 50% from the original thickness of 0.023 mm), the matrix production is not enough to distribute through the textile and create the smoother appearance seen in Fig. 4(g) and Fig. 4(k) when using more solvent (2:1 and 3:1 S/C weight ratios). Using more solvent provides a better opportunity for the film to dissolve completely, supplementing matrix production and allowing the partially dissolved fibres to be coated, contributing to a stronger interface between both reinforcement and matrix, and the textile layers. This strong fibre–matrix interface promotes effective transfer of external loads to the reinforcing fibres [6,45], leading to an increase in modulus as evidenced in Fig. 3 (a). The significant increase in the peel strength using a 3:1 S/C weight ratio, however, highlights the influence of solvent amount on maximising the benefits of using an interleaf film. From Fig. 4(k), it is seen that not only are the fibres smoother, but the pockets of space between interlinking fibres are also filled in more, creating a more consistent surface.

An important factor to consider when processing at elevated temperatures is the tendency for flashing to occur under high pressures, as mentioned previously. When less solvent is applied to the textile, there is a risk that any dissolved cellulose will be removed under compaction, leaving little left to form the matrix. Using more solvent allows any flash expelled from the sides to be compensated for, preventing too much potential matrix to be lost. Whilst the samples produced at 2:1 and 3:1 S/C weight ratios show similar tensile behaviour, the increase in peel strength when using the 3:1 S/C weight ratio suggests that for this process it may be beneficial to opt for extra solvent to ensure sufficient matrix can form under compaction. By using excess solvent, any cellulose lost through flashing is compensated for by increased dissolution, helping to provide enough matrix to promote excellent interlaminar adhesion. Additionally, the matrix can surround the fibres effectively, creating a stronger fibre–matrix interface and a better consolidated ACC which can be seen in Fig. 4(l). Using a 3:1 solvent to cellulose weight ratio provides sufficient solvent to penetrate the cloth stack evenly and yield the most favourable peel strength and modulus without a significant drop in tensile strength. When compared to the sample produced without film, there is a significant increase in Young's modulus and peel strength from 2.18 ± 0.20 GPa to 4.20 ± 0.16 GPa, and 61.72 ± 0.31 N/m to 917.31 ± 2.43 N/m respectively. Interestingly, the composite strength is not affected by the incorporation of the film and therefore less affected by the level of interlayer adhesion. It could therefore be hypothesised that some interlayer delamination occurs before ultimate failure in all these composites and therefore this property is less affected by the interleaf film.

The unprocessed cotton yarns within the textile shown Fig. 4(n) comprise an arrangement of loose smaller fibres that, unbonded, may move about more freely and cause the yarn to expand in the epoxy resin [28]. The overall area of the yarn cross-section will therefore be a combination of individual fibres and the air pockets between them, and on processing these yarns will be compacted under the pressure of the heat press. The loose arrangement of fibres may allow easier penetration of the solvent through the yarns, and any matrix produced from fibre dissolution will help to consolidate the fibres. The resulting decrease in relative size of the yarn cross-section is seen in Fig. 4 across all processed ACCs, suggesting that this is the case. It is worth noting that the values for CSA are unlikely to be strongly related to the amount of fibre dissolved, and more so to their level of consolidation within the samples and 'flattening' under compaction. The measurements from Table 1 suggest that the yarn CSA remains reasonably consistent across all samples, except for the samples produced using a 3:1 S/C ratio and film when it decreases. This reduction is more likely to be a stronger indication of how much the individual fibres have become more closely packed through generation of the fibre–matrix interface. The influence of S/C weight ratio is further emphasised when looking at the calculated densities of the processed ACCs in Table 1, as compared to the expected density of the cellulose materials from known values in literature [41–43]. Density remains relatively consistent across all samples produced using 1:1 and 2:1 S/C weight ratios, considerably lower than the sample produced using a 3:1 S/C weight ratio and interleaf film. The increase in density when using a 3:1 S/C weight ratio supports the notion that using more solvent ensures sufficient matrix can distribute effectively within the fibres and fill the available space between the yarns, and this is reflected in the increased density, aligning more closely with the density of pure cellulose. If a 2:1 S/C weight ratio is used, as the film dissolves, it possibly remains solely between the layers which explain the increase in thickness of the ACC. Although we might expect this to contribute to an increase in peel strength, the lack of sufficient penetration to the fibre assembly prevents this from occurring. Whilst the key aim of the interleaf film is to place matrix in-between textile layers, the importance of filling the space between the interwoven yarns is also important to ensure voids can be reduced. This is also complemented by the difference in yarn size that can be seen across the three S/C weight ratios in Fig. 4(c), Fig. 4(g), and Fig. 4(k). A noticeably smaller yarn size is present when using a 3:1 S/C weight ratio and less void space can be seen within the yarn cross section, suggesting a more consolidated yarn from improved solvent penetration. This particular sample exhibits a potentially optimum combination of matrix and reinforcement, in addition to excellent consolidation of the components. Using a 3:1 S/C weight ratio allows sufficient dissolution, and any loss of cellulose through flashing that occurs under compaction is compensated for, allowing more matrix to remain in the ACC. Whilst voids can still be visibly seen in this sample (Fig. 4(l)), the relative magnitude of these regions compared to the other samples is lower, and improved consolidation of the fibres within the yarns is achieved. The combination of the interleaf film and a sufficient S/C weight ratio allows matrix to be produced and maintained. As more matrix is present, there is sufficient material to fill the pockets of space within the yarns as well as provide more consistent interlaminar matrix to enhance the fibre–matrix interface.

It is important to note that finding the right balance of matrix and fibre within a composite is of equal importance to achieving a strong interface. Both matrix and reinforcement separately contribute their individual properties, and their contribution is typically driven by the respective volume fractions which are present in the composite [46]. There are two possible effects on the resulting fibre volume fraction of the ACC when adding the interleaf film. On one hand, if the film is dissolving instead of the fibre, this may result in less fibre dissolution and a higher volume of fibre present. The interleaf film comprises cellulose II which, due to its comparatively lower degree of polymerisation when compared to native cellulose [47], could be expected to have increased mobility in solution and dissolve faster, indeed more preferentially than the cotton. The method of solvent application, however, ensures that solvent is distributed to both film and textile, allowing it to contact both components equally and initiate dissolution. This leads to the second possibility that adding the film places additional matrix component within the composite, increasing the overall amount of matrix in the ACC. Although the film dissolves, it does not significantly affect the amount of fibre dissolved, that is, the fibre dissolution remains relatively consistent and is independent of the presence of the film. This would result in the same volume of undissolved fibre, but more matrix component, thus reducing the fibre volume fraction, and thus reduce its contribution of properties to the composite. This is evidenced in the decrease in tensile strength observed for the samples with film when compared to the samples made without film, suggesting that the film increases the proportion of matrix component and thus reduces fibre volume fraction. This decrease, however, is relatively small, so it can be seen here that achieving a good balance of matrix and fibre can allow efficient transfer of fibre properties, without excessively reducing fibre volume fraction. Thus, the process of hot compaction and interleaf cellulose film can improve properties overall, without any significant loss of tensile strength. The percentage gain in mass from adding an interleaf film can be calculated by comparing the mass of one layer of film, with the overall mass of two cloth layers. The ratio of film mass to textile mass is 0.1 and so a 10% gain of cellulosic material can be achieved by adding the film during preparation, which effectively ‘dilutes’ the cellulosic reinforcing fibres by 10%. With this, Young’s modulus can be doubled, and furthermore, peel strength can be increased dramatically when using a 3:1 S/C weight ratio, so this is a very effectively strategy for getting an excellent balance of mechanical properties.

3.2. *Influence of interleaf film on crystalline structure*

XRD was used to investigate the changes in the balance of the crystal structure using 3:1 S/C weight ratio with and without the interleaf film. Table 2 summarises the crystalline composition of each sample, as determined by deconvolution of the XRD diffractograms obtained from specimens taken from each sample. A typical diffractogram for each sample is presented in Fig. 5. The deconvolution curves from both samples are presented in Fig. 6.

On addition of the interleaf film, the relative size of the cellulose I peaks, particularly the main reflection at 22.4°, are seen to decrease when compared to the sample prepared without film. At the same time, cellulose II peaks begin to appear more prominently, and the broad amorphous peak at 18.2° increases. As the matrix fraction is a combination of both cellulose II and amorphous cellulose, this supports the earlier suggestion that the increased matrix production brought about by adding the film subsequently reduces the overall fibre volume fraction.

The reduction of cellulose I from 71 ± 1 % to 57 ± 1% confirms the earlier suggested notion that a decrease in fibre volume fraction (which contributes the cellulose I fraction) is brought about by addition of the film and results in a decreased tensile strength. The presence of the film is also highlighted by the increase in cellulose II content, particularly at 21.8°, as compared to the small shoulder seen at this peak for the sample without film. From Fig. 6 this is clearer from the increased size of the peaks at 20.2° and 21.8°, indicative of the presence of cellulose II.

It is worth noting that crystal alignment in a yarn-based architecture is predominately along the vertical axis of a yarn, and so an equatorial diffraction scan of a textile reinforced ACC may result in a 2θ scan where some peaks are absent. Additionally, the helical structure of a yarn can result in some peak modification due to a slight variation in twist angle of fibres within the yarn. Whilst considered, this is not deemed significant in our analysis as the peaks generated in this work have been used simply to highlight the relative change in cellulose I, cellulose II and amorphous cellulose as a result of processing with and without the film. Within these constraints, we believe that this deconvolution technique is a useful method to evaluate the changes in structure and hence follow the dissolution and fractions of the various phases. It also helps to crucially highlight the fact that although the overall relative fraction of the reinforcing fibres is decreased by the incorporation of the film, the modulus increases suggesting that the properties of an ACC (as with all composites) is highly dependent on developing good adhesion between the various phases. The interleaf film offers an excellent method to achieve a good balance of mechanical properties through increase compatibility both within and between the woven textile layers.

Table 2
Crystalline composition of ACCs as analysed by XRD.

	Cellulose I Fraction (%)	Cellulose II Fraction (%)	Amorphous Fraction (%)
ACC - No Film	71 ± 1	6.2 ± 0.2	23 ± 1
ACC -With Film	57 ± 1	8.8 ± 0.4	34 ± 1

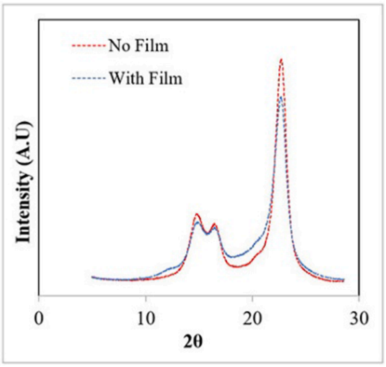

Fig. 5. XRD diffraction patterns of ACCs produced with S/C weight ratios of 3:1, without interleaf film (red line) and with interleaf film (blue line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.3. *Influence of [C2MIM][OAc] % in DMSO*

Viscosities of the solvent solutions used in this work were measured at 25 °C and 100 °C, and the results are shown in Table 3. The addition of DMSO significantly reduces the viscosity of the pure IL solution with a fivefold decrease with the addition of 20 % DMSO. The notable reduction in viscosity as temperature is increased from 25 °C to 100 °C shows that the most likely benefit of adding DMSO is felt mainly at preparation stage, where a significantly reduced viscosity is highly beneficial for applying the S/C solvent to the woven cloth layers. As DMSO wt. % increases, viscosity also decreases but the difference is less apparent at the processing temperature of 100 °C.

Results from mechanical testing are presented in Fig. 7 and images of ACC cross-sections are shown in Fig. 8. Table 4 shows the calculated values for ACC thickness, density, and yarn CSA for ACCs prepared using various wt. % of [C2MIM][OAc] in DMSO.

The importance of achieving a balance of solvent viscosity and sufficient [C2MIM][OAc] is highlighted in Fig. 7 (a), where it can be seen that Young’s modulus decreases as [C2MIM][OAc] % is decreased. This provides some insight into the amount of matrix that has formed within the composite during processing. It is well understood that sufficient matrix formation can support a strong fibre–matrix interface and help to efficiently transfer external loads to the fibres, thus getting the best out of the properties [48]. The highest values for modulus are achieved when using pure, or 80 wt% [C2MIM][OAc], and beyond this, an overall decrease can be observed. As more DMSO is added, viscosity is decreased as highlighted in Table 3. A low viscosity solution would promote excess flashing under compaction, reducing matrix formation that can surround the fibres. Whilst it is beneficial to reduce the viscosity of the solvent to mass transport, the results suggest that it is also

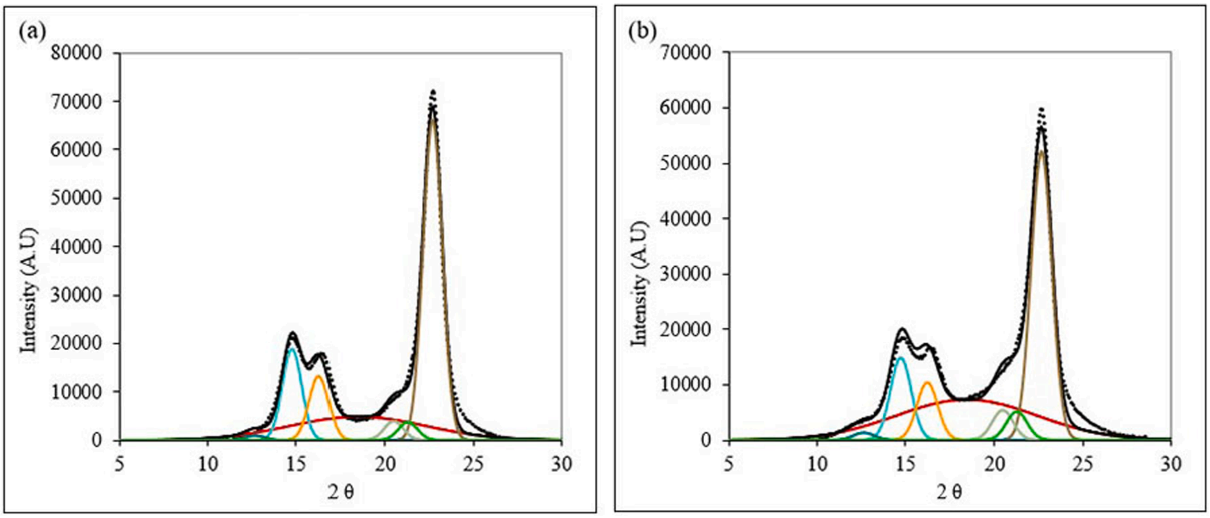

Fig. 6. The deconvolution curves of ACCs produced using a 3:1 solvent to cellulose weight ratio, without interleaf film (a), corresponding to the red curve above and with interleaf film (b) corresponding to the blue curve above. The experimental measurement is shown in the black dotted line, and the black curve is a summation of the crystalline peaks of cellulose I (shown in blue, orange and brown), and cellulose II (shown in grey and green). The amorphous peak is shown in red. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Viscosity measurements of solvent solutions with various wt% [C2MIM][OAc] in DMSO.

% [C2MIM][OAc] in DMSO	Viscosity @ 25 °C (mPa. s)	Viscosity @ 100 °C (mPa. s)
100%	150.1 ± 2.1	7.8 ± 0.9
80%	32.1 ± 2.1	4.68 ± 0.04
70%	28.2 ± 0.2	4.3 ± 0.1
60%	26.6 ± 0.1	3.4 ± 1.0
50%	14.1 ± 2.9	2.6 ± 0.4
40%	10.2 ± 0.5	2.22 ± 0.03
20%	2.8 ± 0.2	0.991 ± 0.001
Pure DMSO	1.5 ± 0.1	0.80 ± 0.04

important not to reduce it to a level that promotes excess flashing. Between 50 and 80 wt% [C2MIM][OAc] there is a consistently high peel strength which is improved from using 100 % pure [C2MIM][OAc]. There is, however, no significant benefit to using more than 20 wt% DMSO as Young's modulus is optimised at 100 wt% and 80 wt% [C2MIM][OAc]. It is interesting to note that when using 50–70 wt% [C2MIM][OAc] in DMSO, peel strength values are still in a range comparable to when using 80 wt% [C2MIM][OAc]. This suggests that there is still sufficient matrix formed even when the amount of [C2MIM][OAc] is significantly reduced. Within a specific time window, the increase in DMSO is potentially speeding up the activity of the [C2MIM][OAc], allowing a relatively smaller concentration to achieve similar dissolution to the larger 80 wt% [C2MIM][OAc] mixture, and this is reflected in the similar peel strength values. It is nonetheless important to consider the balanced of matrix required to support interlaminar adhesion, and the remaining fibre volume fraction required to provide sufficient tensile properties to the material. The decrease in Young's modulus indicates that while sufficient interlaminar adhesion is achieved from dissolution, it not sufficient to enable efficient transfer of external load to the fibre reinforcement. While it can be appreciated that both 100 wt% and 80 wt % of [C2MIM][OAc] in DMSO were seen to yield optimal Young's modulus, the potential cost benefits of offsetting some [C2MIM][OAc]

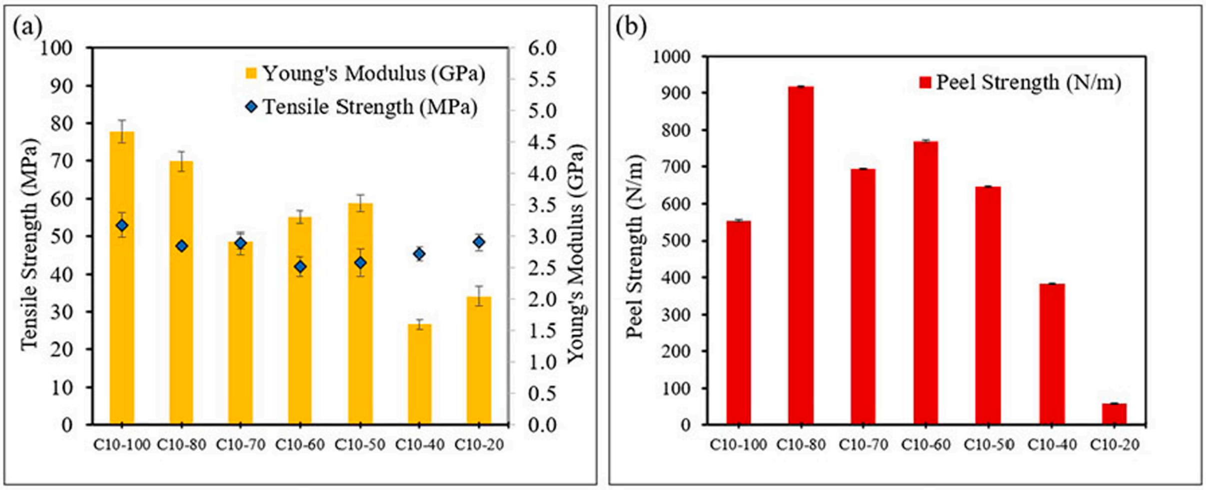

Fig. 7. Mechanical properties of ACCs produced with additional interleaf film in-between layers of cotton textile, using a 3:1 solvent to cellulose weight ratio, using various [C2MIM][OAc] % in DMSO. Tensile strength and Young's modulus are shown in (a) and peel strength is presented in (b). C10-CY corresponds to a sample processed for 10 min using a solvent solution consisting of Y wt. % ionic liquid. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

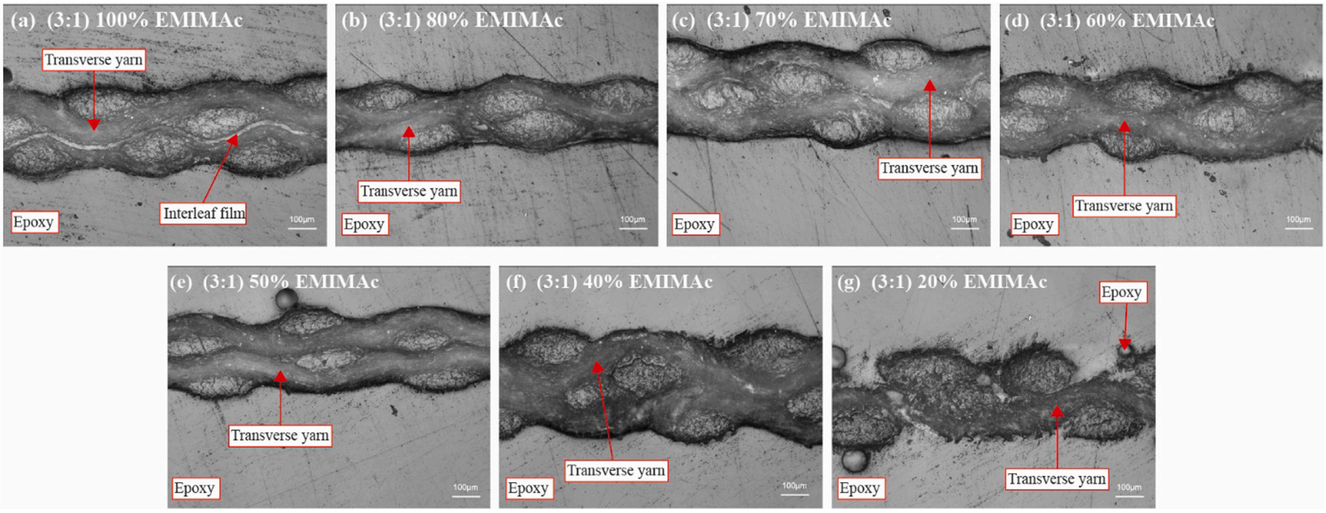

Fig. 8. Cross-sections of prepared ACCs at various weight % [C2MIM][OAc] in DMSO. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 4
Calculated values of thickness (mm), density (g/cc), and yarn CSA (mm2) for ACCs prepared using various weight % of [C2MIM][OAc] in DMSO.

Sample ID	% [C2MIM] [OAc]	Thickness (mm)	Density (g/cc)	Yarn CSA (mm^2)
C10-100	100	0.30 ± 0.01	0.93 ± 0.01	0.021 ± 0.001
C10-80	80	0.22 ± 0.005	1.3 ± 0.02	0.015 ± 0.001
C10-70	70	0.334 ± 0.004	0.95 ± 0.02	0.018 ± 0.002
C10-60	60	0.28 ± 0.02	1.13 ± 0.03	0.019 ± 0.001
C10-50	50	0.27 ± 0.02	0.97 ± 0.03	0.013 ± 0.001
C10-40	40	0.34 ± 0.01	0.86 ± 0.01	0.020 ± 0.004
C10-20	20	0.31 ± 0.01	0.87 ± 0.01	0.025 ± 0.001

with DMSO are worth considering.

Tensile strength remains relatively constant at all concentrations of [C2MIM][OAc]. The narrow range of values, however, suggest the amount of yarn being dissolved does not change significantly, given that tensile strength comes primarily from the reinforcing component of the material [49]. The fibre CSA estimated from image analysis shows a larger variation between values, however, as mentioned earlier the CSA values correlate more to consolidation of the ACCs. Given the visibly thinner appearance of the samples produced using 80 and 60 wt% [C2MIM][OAc] for example, the decrease in visible yarn CSA arguably coincides with the penetration of matrix through the layers, binding the fibres and layers closer together. Conversely, a weaker consolidation is seen when using 20 and 40 wt% [C2MIM][OAc], coinciding with the drop in properties, and density for these samples. The interleaf film can still be seen when pure [C2MIM][OAc] is used, reinforcing the idea that the presence of DMSO supports faster dissolution times and is beneficial when processing for shorter periods. As evidenced in the mechanical properties, however, adding more than 20 wt% DMSO results in less consolidation within the ACC, due to a combination of increased flashing of a less viscous solvent solution and a reduced capacity to dissolve cellulose.

The increased density of a more consolidated sample, coupled with optimised peel strength and modulus suggests that using 80 wt% [C2MIM][OAc] in DMSO with a 3:1 S/C weight ratio is indeed a good combination. A proposed 'optimum' sample using a 3:1 S/C ratio, and 80 wt% [C2MIM][OAc] in DMSO solvent solution was identified for the final phase of the study.

3.4. Influence of interleaf film on out-of-plane properties.

ACC samples were prepared using various stacking arrangements as shown in Fig. 2 following the above optimum solvent configuration. Compaction time, temperature and pressure remained unchanged, at 10 min, 100 °C and 2 MPa respectively. Samples were tested by applying load in three directions relative to the warp yarn orientation: 0° (longitudinal), 90° (transverse), and 45° (bias) directions (Fig. 9). It was hypothesised that the addition of the film may contribute to better stress transfer from the matrix to the yarns, allowing improved properties when subjected to non-axial loading. In addition, a sample of unprocessed cotton cloth was tested, and the results are summarised in Fig. 10 and Fig. 11.

The unprocessed cotton cloth shows the largest variation in tensile properties provided by the warp (longitudinal) and weft (transverse) directions, and this variation is mirrored in the composite prepared without interleaf film. There is a notable increase in Young's modulus in all directions for the ACCs made with the interleaf film, and the difference between longitudinal and transverse properties is reduced considerably. This agrees with the hypothesis that extra matrix formation from the interleaf film helps to stabilise the reinforcement yarns, reducing the effect of crimp on load dissipation and allowing stress to be transferred more efficiently. Although tensile strength reduces through all stacking sequences, it could be argued that this reduction is

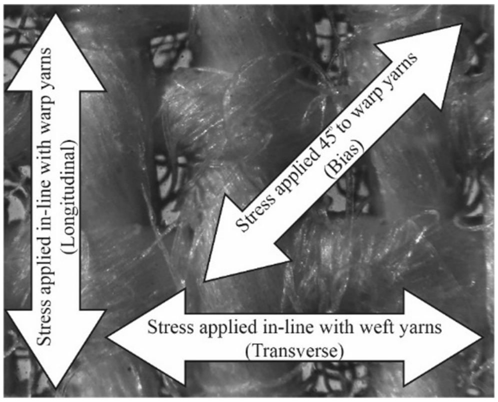

Fig. 9. Direction of longitudinal, transverse, and bias tensile testing of ACCs. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

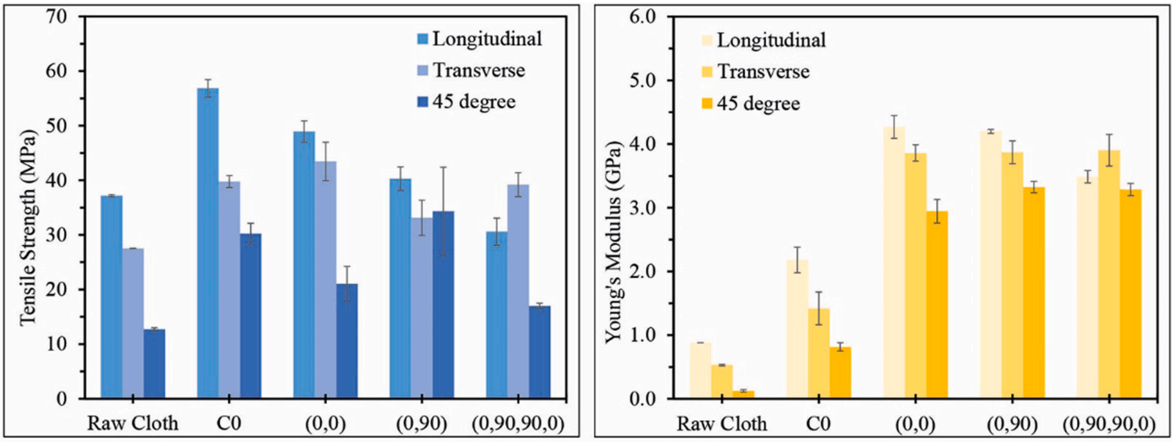

Fig. 10. Comparison of longitudinal (0°), transverse (90°) and bias (45°) mechanical properties of all-cellulose composites prepared with stacking sequences (0,0), (0,90) and (0,90,90,0). Unprocessed cotton cloth (Raw cloth), and the sample made without interleaf film (C0) is also shown for comparison. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

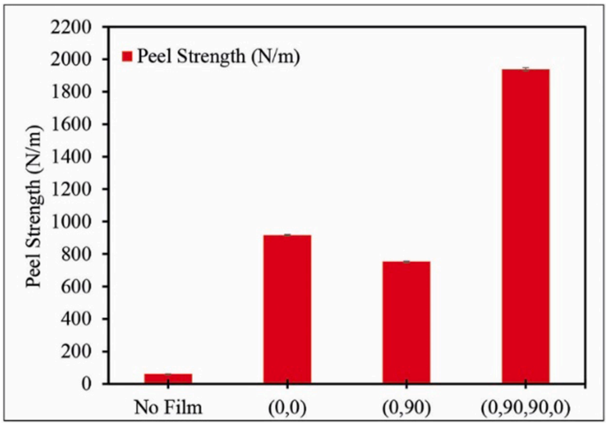

Fig. 11. Comparison of longitudinal peel strength of all-cellulose composites prepared with stacking sequences (0,0), (0,90) and (0,90,90,0). The sample made without interleaf film (C0) is also shown for comparison. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

acceptable when considering the improvement in modulus and peel strength, as seen in Fig. 11.

When using a (0,90) stacking sequence, not only are the properties in the 0°/90° directions brought closer together, but the properties in the 45° bias direction are also increased, in line with classical lamination theory (CLT) [50]. This suggests that the mechanical properties of ACCs follow the behaviour of traditional composite laminates and gives a promising insight into the potential for this method of ACC preparation. Some interesting results and further insights are seen for the symmetric sample produced using a (0,90,90,0) stacking sequence. A considerably large peel strength is obtained with this sample and a minimal variation in modulus in all tested directions, but the notable reduction in tensile strength tells a different story, particularly as in this case, the properties in the 45° direction are considerably reduced. The initial ACCs produced using two cloth layers had one layer of interleaf film, but for the symmetric sample, three interleaf film layers were inserted in between the four cloth layers. This means that when considering the ratio of film mass to textile mass, there is now a 15% increase in cellulosic material from addition of the film, rather than 10% as was the case for the two-layer samples. This would potentially result in more matrix production which is evidenced by the elevated peel strength, but this is achieved at the expense of a lower fibre volume fraction, and a potential shift in failure mode. We earlier hypothesised that for the ACCs produced using two textile layers, there may be some delamination occurring in the ACCs before ultimate failure, when the fibres then break. At this point we suggest that the matrix has already broken, and the majority of the load is taken by the fibres. In this case, whilst there is sufficient interface to promote load transfer from the fibre to the matrix, the interfacial bond is not too strong as to cause the matrix to crack under applied load. For the symmetric sample, the increased matrix production leads to cracks forming in the matrix, which can lead to brittle failure of the composite [51]. This could explain why the off-axis strength of the ACC is lower as the interface becomes compromised as a result of crack propagation. When load is applied in the 45° direction there are no fibres to take the loading, the drop in mechanical properties confirms that the matrix has broken before the fibres.

4. Conclusions

This study has demonstrated the benefits of using interleaf cellulosic films in combination with cotton textiles to prepare all-cellulose composites with an excellent balance of mechanical properties. ACCs were produced via partial dissolution under conditions of temperature and pressure and exhibited excellent interlaminar adhesion brought about by placing extra matrix material where it is needed. The interleaf film dissolves and provides enough matrix where it can have the most effect, to sufficiently bond the textile layers together, allowing the partially dissolved fibres to bond to the interface effectively. By using a 3:1 solvent to cellulose weight ratio, and a solvent solution comprising 80 % [C2MIM][OAc] in 20% wt. DMSO, this method of using interleaved films can obtain a twofold and threefold improvement in Young's modulus and peel strength respectively. The improved adhesion contributes to better stress transfer of external loads, and at a relatively low processing time of 10 min, Young's modulus was significantly improved at the expense of only minimal losses in tensile strength. Furthermore, improvements are seen in the 45° direction when orientating the warp yarns of two layers at 0° and 90°. Given the benefits of combining DMSO with [C2MIM][OAc] have been demonstrated, it would be useful to explore how the addition of DMSO might affect the recycling process of ILs, and this will be included in future research. This novel method shows promise for creating ACCs and offers scope for further exploration into optimising compaction conditions such as time, temperature and pressure. We are currently working on applying design of experiments (DoE) methodology to investigate the interactions between these factors.

CRediT authorship contribution statement

Ashley Victoria: Methodology, Investigation, Formal analysis, Validation, Data curation, Writing – original draft. **Michael Edward Ries:** Supervision, Conceptualization, Methodology, Data curation, Writing – review & editing. **Peter John Hine:** Supervision, Conceptualization, Methodology, Data curation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Availability of data and materials

The datasets generated during and/or analysed during the current study are available at https://doi.org/10.5518/1131

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