



The water absorption behaviour of all-polypropylene composites and its effect on mechanical properties

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ABSTRACT

Woven fabric based self-reinforced polypropylene (SRPP) or all-polypropylene (all-PP) composite laminates based on co-extruded tapes were analysed for water uptake and the effect that this had on mechanical properties. Comparisons were made to a commercial glass mat reinforced PP (GMT) and natural fibre mat reinforced PP (NMT). Although the all-PP composites described in this paper can show significant water uptake through the presence of voids between tapes in the crimp region, the amount of which depends on the fabric architecture and level of consolidation, and their mechanical properties are seen to be largely unaffected by water absorption, even after freezing and thawing. Conversely, the NMT used in this investigation demonstrated significant mechanical degradation with increasing water uptake.

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1. Introduction

1.1. All-polypropylene composites

In recent years, self-reinforced polypropylene composites (SRPP) or all-polypropylene (all-PP) composites have been proposed as an alternative to traditional polypropylene matrix composites for a wide range of applications [1–11]. All-PP composites may have specific ecological advantages over composites based on glass or natural fibres, since they are entirely thermoplastic and can be melted down at the end of the product life without the need for a separate fibre recovery process. Various routes have been proposed so far for the manufacturing of self-reinforced composites [12–21], and two main routes shall be described briefly here. The ‘hot-compaction’ process was initially developed at the University of Leeds as a route to self-reinforced polyethylene although it has also been applied to other polymers [15,22–25]. This hot-compaction method selectively melts the surface of (mono-component) polymer fibres and welds them together to form a composite of retained fibres embedded in a melted and

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moduli of >6 GPa and tensile strengths of >200 MPa [27], which makes them an interesting alternative to replace traditional engineering composites, such as natural or glass fibre reinforced PP [36–40], for a wide range of applications, notably those requiring high impact resistance [41].

The co-extruded PP tapes used for the manufacture of SRPP or all-PP composites are drawn in the solid state to very high draw ratios (typically $\lambda \sim 17$), which leads to so-called ‘over-drawing’ [42,43]. This overdrawing is associated with a decrease in density and a reduction in transparency due to the creation of micro-voiding within the tapes. Tapes of a draw ratio of 17 have a density is 0.73 g cm^{-3} , which is almost 20% below that of non-voided tapes of lower draw ratio ($\lambda < 9$) [32,43]. Tapes with a draw ratio of 17 are also optically opaque because the presence of the microvoids leads to light scattering. Although these voids reduce composite density and so are attractive for producing laminates with high specific mechanical properties [27], it is important to consider any possible negative effects. It is believed that these “intra-tape” voids are closed, but it is possible that they could lead to an additional mechanism for liquid absorption.

Alcock et al. [27] demonstrated that, depending on the compaction conditions (pressure and temperature), all-PP composites can also possess varying amounts of voids between adjacent fabric plies in the consolidated composite structure. Although the use of PP tapes rather than PP fibres reduces the degree of crimping, inter-tape voids still occur between adjacent fabric plies. These inter-tape voids result from incomplete consolidation, may also be a possible route for water uptake. The combination of intra-tape and inter-tape voiding may affect the environmental performance of all-PP composites since these voids may be a route for fluid ingress into the composite structure.

1.2. Water absorption of composites

The way in which composite materials absorb water depends upon many factors, such as temperature, fibre volume fraction, fibre orientation, fibre type, area of exposed surfaces, interfacial bonding, diffusivity, reaction between water and matrix, surface protection and voids [37,40,44–49].

Different models have been proposed to describe the moisture absorption of materials [44], which can also predict the temperature and moisture distribution inside a material. This is known as a ‘moisture problem’, and may be solved analytically by Fickian diffusion. When the moisture absorption curve fits the linear Fickian diffusion curve, as shown in Fig. 1, the moisture absorption process can be considered as Fickian diffusion. Subsequently, the maximum moisture content and mass diffusivity can be obtained from Eq. (1).

$$\frac{M_t}{M_m} = 1 - \frac{8}{\pi^2} \sum_{j=0}^{\infty} \frac{1}{(2j+1)^2} e^{-D(2j+1)^2 \pi^2 t/h^2} \quad (1)$$

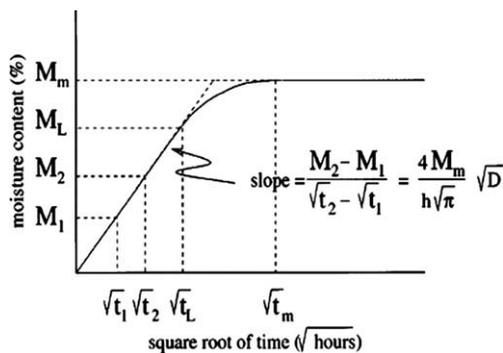


Fig. 1. Illustration of the change of moisture content with the square root of time for Fickian diffusion. For $t < t_L$ the slope is constant [48].

where M_t is the mass of the composites after immersion time, t . D is the mass diffusivity, M_m is the maximum moisture content in the material, and h is the thickness of the materials.

Some linear Fickian diffusion processes for composites were reported by Springer [44]. Nevertheless, many of the processes reported were determined to be non-linear Fickian diffusion processes and had multi-stage moisture absorption processes. The reason for this is likely to be that several different moisture absorption processes may be active in a single material.

The purpose of this paper is to investigate the water absorption behaviour of all-PP composites based on consolidated fabrics of woven co-extruded PP tapes, and the effect of water absorption on mechanical properties. Since PP is oleophilic, the oil absorption capacity of all-PP composites is also presented in this paper. A comparison is made between the water absorption behaviour of all-PP composites, commercial glass mat reinforced thermoplastics composites (GMT) and natural fibre mat reinforced thermoplastic composites (NMT), the latter being known to be particularly susceptible to moisture absorption [37,40,44].

2. Experimental

2.1. Materials and processing

The co-extruded tape used to make the all-PP composites described in this paper was manufactured by Lankhorst Indutech BV (now Lankhorst Pure Composites, The Netherlands), and then woven into plain weave fabric by BW Industrial (The Netherlands). The density of the co-extruded tape is 0.73 g cm^{-3} , while the areal density of the all-PP fabric is 103 g m^{-2} [27].

It has been shown previously that all-PP composites can be consolidated inside a “process feasibility window” of varying temperature and pressure [27,32,34,41], and the mechanical properties and the degree of consolidation of the laminate depend strongly on the processing parameters used. To investigate the influence of the degree of consolidation of the all-PP laminates on moisture absorption, two sets of all-PP laminates were manufactured. One set of all-PP laminates was compacted at a higher consolidation pressure (4 MPa), while another set was consolidated at a lower consolidation pressure (0.1 MPa). The full process for the manufacture of all-PP laminates by either a hot-pressing or vacuum bagging route has been described in full detail previously [27]. The first set of all-PP composites were processed in a hot-press by consolidating a stack of 24 layers of all-PP fabric at a temperature of $145 \text{ }^\circ\text{C}$ and a pressure of 4 MPa. The second set of all-PP composites was also consolidated at a temperature of $145 \text{ }^\circ\text{C}$ by using a vacuum bagging technique [27,34]. In this process, a stack of 24 plies of all-PP fabric is sealed in a vacuum bag which is then placed inside a cold oven. The vacuum bag is connected to a vacuum pump that applies a negative pressure inside the vacuum bag, and so atmospheric pressure ($\sim 0.1 \text{ MPa}$) is used to compact the laminate while the oven is heated to the consolidation temperature.

In this paper, the “edges” of the laminates are defined as the four surfaces of the laminates normal to the laminate plane, i.e. when test specimens are cut from a larger laminate, while the “faces” of the laminate are the two surfaces in parallel to the plane of the laminate. To investigate the influence of water absorption through the faces or the edges of the composite laminates, an additional set of all-PP laminates were produced in a hot-press (at a temperature of $145 \text{ }^\circ\text{C}$ and a pressure of 4 MPa) from 24 layers of all-PP fabric. During stacking of the all-PP fabric plies between the platens of the hot press, this additional set had a $\sim 30 \text{ }\mu\text{m}$ thick PP film laminated onto the top and bottom faces. This was performed to attempt to seal the top and bottom faces of the laminate so that any subsequent water uptake would only occur through the edges of the laminates.

In addition to all-PP composites, conventional commercial PP matrix composites were also investigated. GMT and NMT are short random fibre reinforced polypropylene composites, supplied by Quadrant (Switzerland). The glass and flax fibre fractions are both 30 wt.%. The thicknesses of the GMT and NMT sheets were 1.7 mm and 4.5 mm, respectively. To directly compare the mechanical properties of all-PP laminates with GMT, all-PP laminates were also produced from 12 layers of woven tape fabrics (total thickness 1.7 mm) compacted by means of a continuous double-belt press (145 °C and 2.5 MPa) at the Institut für Verbundwerkstoffe GmbH (Kaiserslautern, Germany).

2.2. Water absorption tests

Specimens used for the water absorption tests were cut from 24-ply all-PP laminates (processed at both 0.1 and 4 MPa) with dimensions in accordance with ASTM D570 (77 × 25 × 3.2 mm). For comparison, GMT and NMT composites and samples of a ethylene-propylene copolymer (as was used to provide the skin layer of the tapes during co-extrusion [32]) were also tested. At least three specimens of each material were tested. All specimens were dried in an oven for 24 h at 60 °C and weighed before immersion in a distilled water bath at different temperatures; 23 °C (room temperature), 40 °C and 65 °C. The specimens were periodically removed from the water bath, wiped free of surface moisture, immediately weighed to the nearest 1 mg using a Mettler AE200 balance and then replaced into the water bath.

In order to determine the effect of water uptake on the mechanical properties of the composites, different specimen dimensions were required. Specimens were cut from the belt-pressed (12-ply) all-PP laminates, GMT and NMT sheets into 210 × 15 mm rectangular bars. Three specimens from each material were taken after immersion in water for 40 days at 65 °C and frozen at –18 °C for 24 h. These specimens were then thawed and tested in tension to determine if the expansion of the absorbed water during freezing had affected the tensile performance of the composites.

2.3. Oil absorption tests

The same method as used for water absorption could not be used for oil absorption, as oil cannot be efficiently wiped off prior to weighing. Instead, oil absorption was determined by using 24-ply all-PP laminates consolidated at 145 °C at 4 MPa pressure in a hot press, with and without PP copolymer sheets on the laminate faces. The oil absorption was determined by measuring the change in buoyant force. According to the Archimedes principle, the buoyant force of a solid less dense than the fluid in which it is immersed, is equal to the weight of the volume of fluid it displaces when submerged, less the dry weight of the object. Fluid absorbed by the solid lowers the buoyant force by reducing the volume of

fluid actually displaced. Hence as the buoyant force decreases, the apparent weight of the solid in the oil, m_{oil} , increases and this variation of weight of the immersed solid is equal to the weight of the oil absorbed. The weight of oil absorbed can be practically determined by using a standard density measuring apparatus attached to a microbalance. The oil absorbed in weight percent is derived from the following expression:

$$\text{Oil absorbed (\% w/w) at time } (t) = \frac{m_{oil(t)} - m_{oil(t=0)}}{m_{air}} \quad (2)$$

Where $m_{oil(t=0)}$ is the apparent weight of the specimen when first immersed in oil, $m_{oil(t)}$ is the apparent weight of the specimen in oil after time, t , and m_{air} is the weight of the specimen in air. The oil chosen for the test was a hydraulic oil type AW ISO 32 supplied by Enerpac (HF-95).

2.4. Mechanical testing

Tensile tests were conducted in an Instron 6025 series universal testing machine using a 30kN load cell with data acquisition software. Accurate strain measurements were performed using a clip-on extensometer. All soaked and un-soaked specimens were cut into rectangular tensile specimens with dimensions of 210 × 15 mm, and a gauge length of 110 mm and a test speed of 5 mm/min were used. The Young's moduli were calculated as the gradient of the stress vs. strain curve in a strain range of 0.02–0.40%. Specimens that were considered to have failed in the gripped region were discarded. At least three specimens were tested for each condition.

3. Results and discussion

3.1. The presence of inter-tape and intra-tape voids in all-PP composites

Fig. 2a and b shows typical microstructures of all-PP composite laminates consolidated by vacuum bagging at 0.1 MPa and by hot pressing at 4 MPa respectively. In Fig. 2, the light areas are tapes travelling along the width of the specimen from left to right on the page. The dark areas are tapes travelling out of the plane of the page. In cross-sections of all-PP composite laminates processed by vacuum bagging at low pressure (0.1 MPa), large voids are clearly present in the laminate. In cross sections of all-PP composite laminates processed by hot pressing at a high pressure (4 MPa), it can be seen that most of the inter-tape voids disappear but some voids at the level of the fabric crimp of the woven laminates still remain. For higher consolidation pressures, the density of the laminates can be higher than that of the original co-extruded tape (0.73 g cm⁻³) [26]. A possible reason for this is that intrinsic intra-tape voiding that occur in the tape during 'overdrawing' at

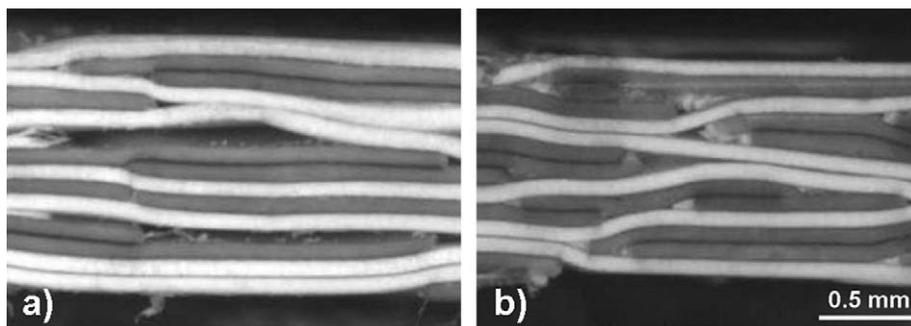


Fig. 2. (a) All-PP woven fabric composite compacted at 0.1 MPa pressure at 145 °C and (b) 4 MPa pressure at 145 °C. Inter-tape voids can still be seen between adjacent tapes in the crimp region even at high pressure consolidation [27].

relatively high draw ratios may be partially closed by the high compressive stresses [42].

Since inter-tape voids in all-PP composites based on co-extruded tapes mainly appear in the region of the crimp in woven laminates, it is expected that fibre architecture in the fabrics can play a significant role in the void content. For example, twill-, satin- or non-crimp fabrics would most likely all lead to lower void contents than the plain weave fabrics used here. For comparison, a cross-section of a unidirectional all-PP composite is shown in Fig. 3 which is fabricated by winding tape around a steel plate and then consolidated in a hot press [26], and the void free nature of the non-crimp structure is clearly visible.

As mentioned above, in addition to the more obvious inter-tape voids, all-PP laminates can also exhibit intra-tape voids. The co-extruded PP tapes used here are drawn in the solid state to very high draw ratios (typically $\lambda = 17$), which leads to a decrease in density and a change in transparency of the tape [42,43,50]. This is due to so-called overdrawing creating micro-voiding within the tape [42]. As described earlier, at $\lambda = 17$, the density of the PP tapes is 0.73 g cm^{-3} , which is almost 20% below that of non-voided tapes at lower draw ratio ($\lambda < 9$). This large decrease in density was attributed to the presence of voids within the tape. Fig. 4 shows, after permanganic etching of the tape, a regular alignment of holes parallel to the drawing direction. Previous confocal laser scanning microscopy investigations have shown that these morphological features are not just limited to the surface but extend into bulk of the tape [42].

3.2. Water absorption of all-PP, GMT and NMT

The water absorption behaviour of all-polypropylene composites compacted at either 0.1 MPa or 4 MPa pressure is compared

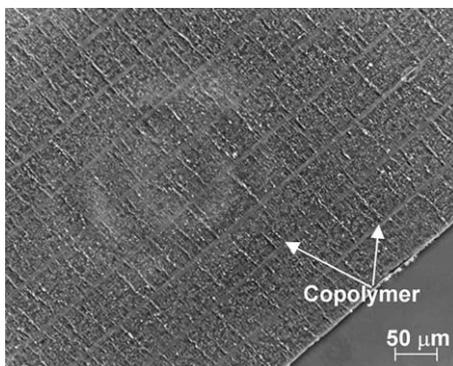


Fig. 3. Optical micrograph under transmitted light of the microtomed cross-section of a compacted unidirectional specimen [50].

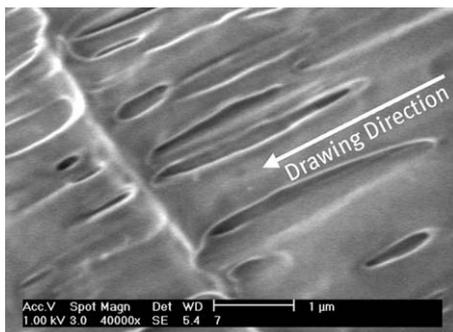


Fig. 4. ESEM of drawn all-PP tape after permanganate etching showing intra-tape voids [42].

to other polypropylene matrix composites (GMT and NMT) and in samples of isotropic PP copolymer of the same grade as used for the skin layer of the co-extruded tapes. Fig. 5 shows the weight fraction of water absorbed by these materials after 1-month immersion in distilled water, normalised for the weight of the composite specimen. Polypropylene is a hydrophobic polymer, and this is reflected in the negligible amount of water absorbed by the isotropic PP copolymer sample (0.03% w/w). The introduction of reinforcing fibres in GMT or NMT increases the sensitivity of PP towards water and the effect is more pronounced for the flax fibre based NMT composite (5.5% w/w) than for the GMT material (0.6% w/w). Concerning all-PP composites, the density of the composite laminate was seen to control the degree of water absorption. The laminate consolidated by vacuum bagging at 0.1 MPa, exhibiting the lowest density and therefore highest void content, absorbs much more water (22% w/w) than the laminate compacted by hot pressing at 4 MPa, which absorbs 4.5% w/w. The presence of the voids in the laminate (as shown in Fig. 2) may explain this behaviour.

Attempts were made to define whether water is also retained in the internal microvoids of the individual tapes, but the low linear mass of the tape (around 1000 denier) did not allow any reliable measurement of water uptake of a single tape. However, since the intra-tape voids, or regions of reduced density, within the PP tape are sealed on both faces by the copolymer skin layers, a barrier is formed between the environment and the highly drawn core layer, and so it was assumed that water absorbed within the tape structure can be neglected.

The laminate geometry may determine a different rate of absorption via the edges compared to the faces of a laminate. In order to characterise the relative importance of these surfaces, water absorption of all-PP composite laminates with and without the addition of isotropic surface films were tested. All-PP composites were consolidated by hot pressing at 4 MPa and were laminated on both faces with copolymer films to prevent water absorption through the top and bottom faces of the panel. The edges account only for 15% of the total surface area of the specimen. Fig. 6 presents the water absorption as a function of time for the two different laminates. Initially the laminate without the surface films absorbs slightly more water but the absorption profiles are not significantly different, which implies that absorption is dominated by the edges of the panel. Only the first 11 days of the water absorption profile are plotted in Fig. 6. After one month, the laminates reached similar water absorption levels: $4.4 \pm 0.2\%$ (without surface films) and $4.7 \pm 0.3\%$ (with surface films). Because water absorption through the edges is dominating, the diffusion coefficient was not evaluated [37].

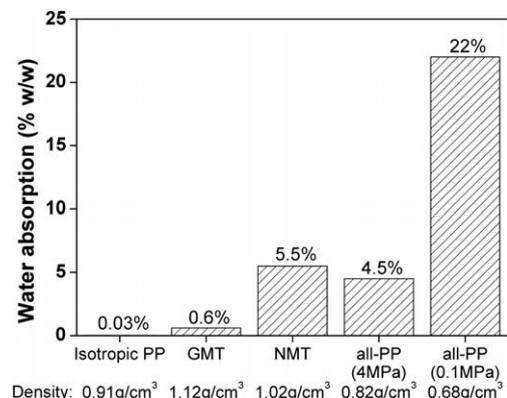


Fig. 5. Weight fraction of water absorbed by various polypropylene composites after 1-month immersion in distilled water. All-PP composites are suffixed by the pressure used to consolidate the laminates (4 MPa by hot pressing or 0.1 MPa by vacuum bagging routes).

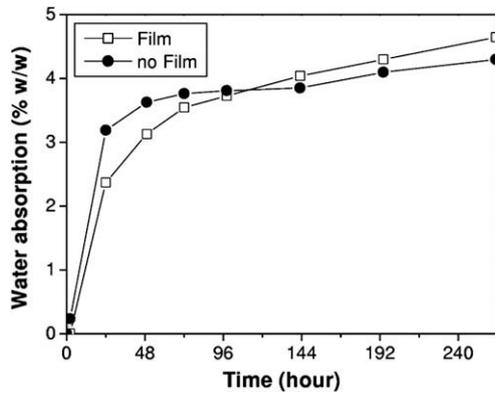


Fig. 6. Water absorption as a function of time of all-PP compacted at 145 °C and 4 MPa pressure. The top and bottom faces of one specimen were sealed by 30 μm thick PP films. Note the similarity between the two laminates indicating that water absorption mainly occurs through the edges.

3.3. Oil absorption of all-PP

Although polypropylene is hydrophobic, it is oleophilic and the oil sorption capacity of polypropylene allows polypropylene fibres to be used as a synthetic sorbent material, for example in the case of oil spills in open water [46]. The oil absorption of an all-PP laminate compacted by hot pressing at 145 °C and 4 MPa is presented in Fig. 7. The oil absorption was measured by buoyancy as described earlier, and the oil density at 25 °C was measured to be 0.87 g cm^{-3} . The density of the laminate could then be determined by measuring the weight of the specimen, and applying Eq. (3).

$$\rho_{\text{solid}} = \rho_{\text{oil}} \times \left(\frac{m_{\text{air}}}{m_{\text{air}} - m_{\text{oil}}} \right) \quad (3)$$

Where ρ_{solid} and ρ_{oil} are the densities of the solid immersed body and the oil respectively, and m_{air} and m_{oil} are the masses of the solid when measured in air and in oil respectively. During the third day of immersion, the specimen started to sink. As oil replaces air in the porous material, the density of the laminate increases and eventually becomes greater than that of the oil. By comparing Figs. 6 and 7, it can be seen that a higher weight fraction of oil is absorbed than water in spite of the lower density of the oil. This result suggests that oil is also retained within the tapes unlike in the case of water. This hypothesis was easily confirmed by dropping 100 ml of oil on a ply of unconsolidated all-PP fabric, as shown in Fig. 8. When the tape absorbs oil and the micro-cavities are filled, its refractive index is changed so that light is not scattered and the tapes become fully transparent.

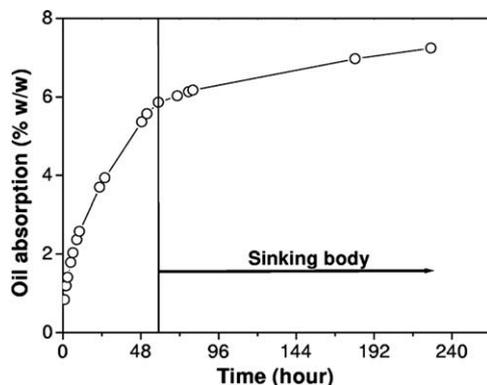


Fig. 7. Oil absorption as a function of time. During the third day of immersion in oil, the density of the all-PP specimen is higher than that of the oil and the specimen sinks.

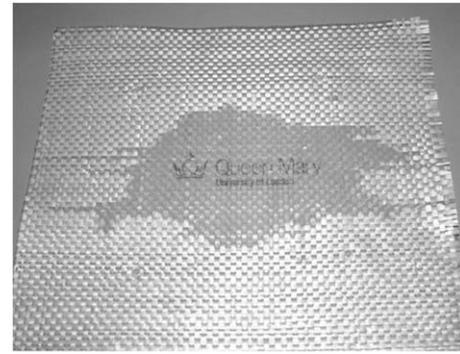


Fig. 8. Oil absorption by woven all-PP tapes. The normally opaque tapes become transparent when in contact to a small volume of oil in the centre of the fabric. This change in optical properties is thought to be due to oil filling the voids within the PP tapes, reducing the amount of light scattered by the voids.

3.4. Influence of water absorption on mechanical properties

One of the main objectives of this investigation was to study the effect of water absorption on the mechanical properties of the composites. For the purpose of this study, some level of voids in the all-PP composites is actually desirable since this allows the evaluation of the effect of these voids on any subsequent reduction in mechanical properties. Thus all-PP composite laminates were not processed at optimised conditions, but instead consolidated by belt pressing at conditions that would provide some degree of inter-tape voiding. It should be noted that compaction at higher temperatures and pressures may well lead to lower void levels [27], and reduce any negative effects of water absorption on mechanical properties.

Figs. 9–11 present the water absorption at different water temperatures as a function of the square root of time for double-belt pressed all-PP (processed at 2.5 MPa pressure and 145 °C), GMT and NMT, respectively. All specimens have the same surface dimension (210 mm \times 15 mm) and unsealed edges. All materials take up water at a greater rate at elevated temperatures. Furthermore, all-PP specimens (Fig. 9) show a clear multi-stage absorption process that does not display Fickian behaviour (as illustrated in Fig. 1). In the case of all-PP specimens, water is thought to enter the laminate through the channels in the edges, first at a lower rate and then more quickly, and also depending on water temperature.

Conversely, the GMT and NMT specimens show moisture absorption behaviour that appears to conform to a typical Fickian diffusion curve (Figs. 10 and 11). In the case of NMT the moisture content is linear with the square root of time until about 40 days

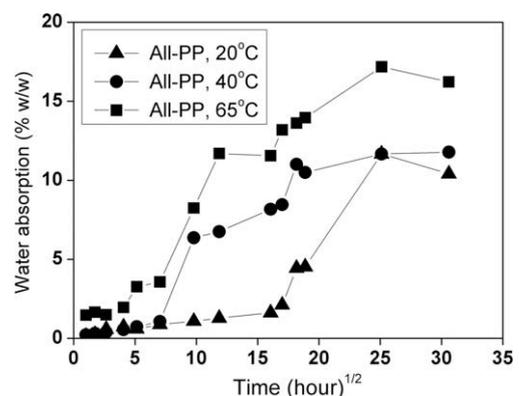


Fig. 9. Water absorption curve for all-PP specimen as a function of square root of time.

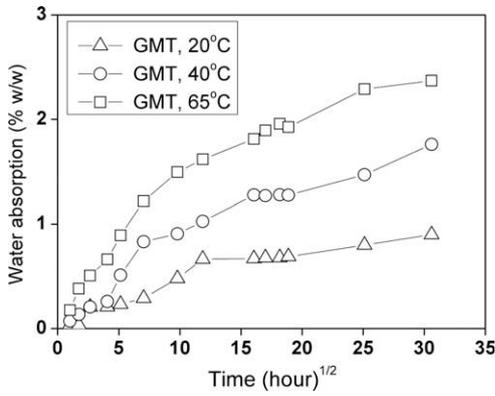


Fig. 10. Water absorption curve for GMT specimen as a function of square root of time.

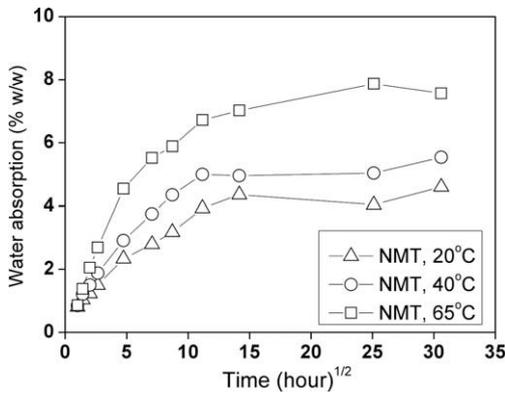


Fig. 11. Water absorption curve for NMT specimen as a function of square root of time.

immersion in water. After that, the moisture content appears to reach saturation.

All specimens used for the water absorption tests were subsequently loaded to failure in tension. The tensile strength and moduli of the all-PP, GMT and NMT composite laminates, are presented in Figs. 12–14 respectively. Five different types of specimens are compared in these tests, and are tested dry (as received/produced), tested after immersion in water at 20 °C, 40 °C or 65 °C, or tested after immersion in water and subsequent freezing and thawing. Specifically in Figs. 12–14, the samples are labelled: ‘dry’ which are specimens that have not been immersed in water, ‘20’, ‘40’ or

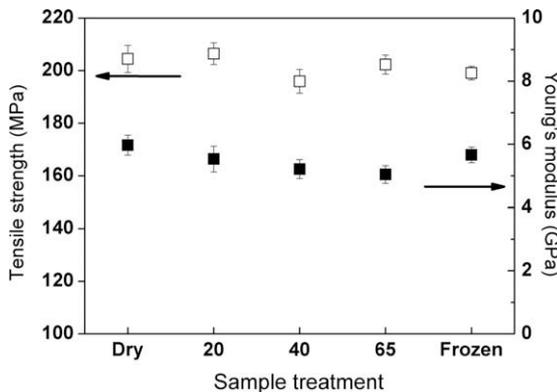


Fig. 12. Mechanical properties of all-PP composites before (dry) and after nearly 40 days immersion in water at different temperature (20 °C, 40 °C and 65 °C) and subsequently freezing.

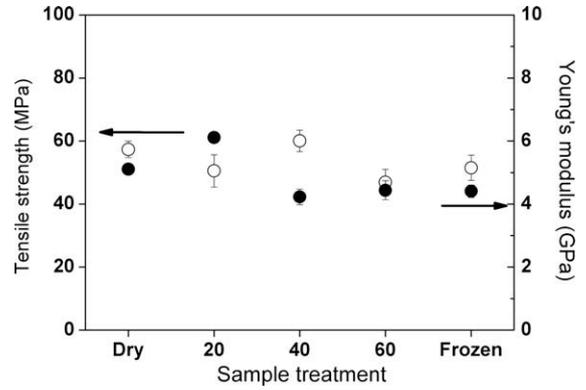


Fig. 13. Mechanical properties of GMT composites before (dry) and after nearly 40 days immersion in water at different temperature (20 °C, 40 °C and 65 °C) and subsequently freezing.

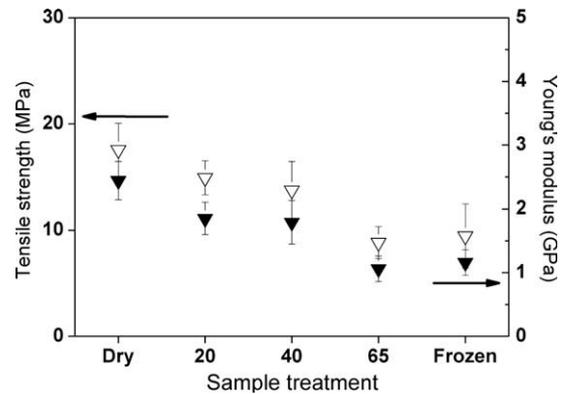


Fig. 14. Mechanical properties of NMT composites before (dry) and after nearly 40 days immersion in water at different temperature (20 °C, 40 °C and 65 °C) and subsequently freezing.

‘65’ which are specimens which have been immersed in water at different temperatures (20 °C, 40 °C and 65 °C, respectively) for approximately 1000 h (~40 days), or ‘Frozen’, which are specimens that have been immersed in water at 65 °C for 40 days, and subsequently frozen at –18 °C for 24 h and then thawed.

Fig. 12 presents the tensile strengths and moduli of all-PP composite laminates. The mechanical properties show negligible reduction due to water absorption. Moreover, they showed no loss in strength and stiffness after freezing at –18 °C for 24 h and subsequent thawing. This is probably due to the high toughness of all-PP composites, even at low temperatures [28,29]. The effect of freezing and thawing was evaluated since the expansion which occurs due to water freezing might lead to micro-cracking and a resultant decrease in mechanical properties of the laminates. Although all tests were performed at room temperature, it was thought that the absence of a strong glass transition temperature in all-PP composites would mean that mechanical properties would not be greatly different if tested while still frozen at –18 °C. In fact, previous studies have shown that all-PP composites show slightly greater tensile strength and tensile stiffness if tested in these low temperature ranges [28].

GMT laminates are not significantly affected by immersion in water, as shown by the relatively low water uptake presented in Fig. 10, and the small changes in mechanical properties presented in Fig. 13. However, in the case of NMT the mechanical properties have changed significantly with water absorption, as shown in Fig. 14. Previous studies into the mechanisms of reduction in mechanical properties of natural fibre composites have shown that

this degradation is most likely the result of fibre degradation rather than interface failure or matrix cracking [37,40]. Although a decrease in mechanical properties was seen after water immersion, no further decrease was seen as a consequence of freezing and thawing. Unlike composite systems based on brittle thermosetting resins such as unsaturated polyester, micro-cracking of the matrix as a result of fibre swelling and freezing is not likely to occur in the case of a ductile matrix such as PP.

4. Conclusion

It can be concluded that woven fabric all-PP composites described in this paper absorb water in spite of the hydrophobic character of polypropylene. The rate of water absorption is mainly dominated by the edges of the laminate and the effect may be minimised if the laminate is well compacted, reducing the amount of inter-ply voids. Because the water absorption route for all-PP composite laminates appears to be by inter-tape voids between tapes that are accessible through the edges, these materials do not follow Fickian diffusion behaviour. The NMT and GMT composites investigated here, on the other hand, display near-Fickian behaviour. For the case of all-PP composite laminates, it is believed that water is mainly retained in the (open) voids between tapes and not in the internal (closed) voids within the tape itself. However, this is not the case if oil is brought into contact with these over-drawn PP tapes. Here, tapes in unconsolidated fabric plies even become transparent as the oil fills the internal voids within the tape, reducing the light scattering by the voids.

A negligible loss in tensile strength and Young's modulus due to the ingress of water was observed for all-PP and GMT composite laminates. Even after freezing and subsequent thawing no changes in mechanical performance were found for these materials, when compared to materials which had been immersed in water but not frozen. However, significant reductions in strength and stiffness were observed for NMT laminates with water uptake.

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References

- [1] Abo El-Maaty MI, Bassett DC, Olley RH, Hine PJ, Ward IM. The hot compaction of polypropylene fibres. *J Mater Sci* 1996;31:1157–63.
- [2] Hine PJ, Ward IM, Teckoe J. The hot compaction of woven polypropylene tapes. *J Mater Sci* 1998;33:2725–33.
- [3] Loos J, Schimanski T, Hofman J, Peijs T, Lemstra PJ. Morphological investigation of polypropylene single-fibre reinforced polypropylene model composites. *Polymer* 2001;42:3827–34.
- [4] Barkoula N-M, Schimanski T, Loos J, Peijs T. Processing of single polymer composites using the concept of constrained fibers. *Polym Compos* 2004;26(1):114–20.
- [5] Ward IM, Hine PJ. The science and technology of hot compaction. *Polymer* 2004;45:1413–27.
- [6] Cabrera N, Alcock B, Loos J, Peijs T. Processing of all-polypropylene composites for ultimate recyclability. In: Proceedings of the institute of mechanical engineers. *J Mater: Des Appl* 2004; 218(Part L2): 145–56.
- [7] Bárány T, Izer A, Czigány T. On consolidation of self-reinforced polypropylene composites. *Plast Rubber Compos* 2006;35(9):375–9.
- [8] Izer A, Bárány T. Hot consolidated all-PP composites from textile fabrics composed of isotactic PP filaments with different degrees of orientation. *Express Polym Lett* 2007;12:790–6.
- [9] McKown S, Cantwell WJ. Investigation of strain-rate effects in self-reinforced polypropylene composites. *J Compos Mater* 2007;41(20):2457–70.
- [10] Banik K, Karger-Kocsis J, Abraham T. Flexural creep of all-polypropylene composites: model analysis. *Polym Eng Sci* 2008;48:941–8.
- [11] Kim KJ, Yu W-R, Harrison P. Optimum consolidation of self-reinforced polypropylene composite and its time-dependent deformation behavior. *Composites Part A* 2008;39(10):1597–605.
- [12] Capiati NJ, Porter RS. The concept of one polymer composites modelled with high density polyethylenes. *J Mater Sci* 1975;10:1671–7.
- [13] Mead TW, Porter RS. The preparation and tensile properties of polyethylene composites. *J Appl Polym Sci* 1978;22:3249–65.
- [14] Chand N, Kreuzberger S, Hinrichsen G. Influence of processing conditions on the tensile properties of unidirectional UHMPE fibre/LDPE composites. *Composites* 1993;25(9):878–90.
- [15] Hine PJ, Ward IM, Olley RH, Bassett DC. The hot compaction of high modulus melt-spun polyethylene fibres. *J Mater Sci* 1993;28:316–24.
- [16] Lacroix F, Lu H-Q, Schulte K. Wet powder impregnation for polyethylene composites: preparation and mechanical properties. *Composites Part A* 1999;30:369–73.
- [17] Pegoretti A, Zanolli A, Migliaresi C. Flexural and interlaminar mechanical properties of unidirectional liquid crystalline single-polymer composites. *Compos Sci Technol* 2006;66(13):1953–62.
- [18] Pegoretti A, Zanolli A, Migliaresi C. Preparation and tensile mechanical properties of unidirectional liquid crystalline single-polymer composites. *Compos Sci Technol* 2006;66(13):1970–9.
- [19] Abraham TN, Siengchin S, Karger-Kocsis J. Dynamic mechanical thermal analysis of all-PP composites based on α and β polymorphic forms. *J Mater Sci* 2008;43(10):3697–703.
- [20] Zhang JM, Reynolds CT, Peijs T. All-poly(ethylene terephthalate) composites by film stacking of oriented tapes. *Composites Part A* 2009;40(11):1747–55.
- [21] Bárány T, Izer A, Karger-Kocsis J. Impact resistance of all-polypropylene composites composed of α and β modifications. *Polym Test* 2009;28(1):76–82.
- [22] Rasburn J, Hine PJ, Ward IM, Olley RH, Bassett DC, Kabeel MA. The hot compaction of polyethylene terephthalate. *J Mater Sci* 1995(30):615–22.
- [23] Hine PJ, Bonner M, Brew B, Ward IM. Hot compacted polypropylene sheet. *Plast Rubber Compos Process Appl* 1998;27(4):167–71.
- [24] Hine PJ, Astruc A, Ward IM. Hot compaction of polyethylene naphthalate. *J Appl Polym Sci* 2004;93(2):796–802.
- [25] Hine PJ, Ward IM. Hot compaction of woven nylon 6, 6 multifilaments. *J Appl Polym Sci* 2006;101(2):991–7.
- [26] Alcock B, Cabrera NO, Barkoula N-M, Loos J, Peijs T. The mechanical properties of unidirectional all-polypropylene composites. *Composites Part A* 2006;37(5):716–26.
- [27] Alcock B, Cabrera NO, Barkoula N-M, Spoelstra AB, Loos J, Peijs T. The mechanical properties of woven tape all-polypropylene composites. *Composites Part A* 2007;38(1):147–61.
- [28] Alcock B, Cabrera NO, Barkoula N-M, Reynolds CT, Govaert LE, Peijs T. The effect of temperature and strain rate on the mechanical properties of highly oriented polypropylene tapes and all-polypropylene composites. *Compos Sci Technol* 2007;67(10):2061–70.
- [29] Alcock B, Cabrera NO, Barkoula N-M, Wang Z, Peijs T. The effect of temperature and strain rate on the impact performance of recyclable all-polypropylene composites. *Composites Part B* 2008;39(3):537–47.
- [30] Barkoula N-M, Alcock B, Cabrera N, Peijs T. Fatigue properties of highly oriented polypropylene tapes and all-polypropylene composites. *Polym Polym Compos* 2008;16(2):101–13.
- [31] Cabrera N, Alcock B, Klompen ETJ, Peijs T. Filament winding of co-extruded polypropylene tapes for fully recyclable all-polypropylene composite products. *Appl Compos Mater* 2008;15(1):27–45.
- [32] Alcock B, Cabrera NO, Barkoula N-M, Loos J, Peijs T. Interfacial properties of highly oriented coextruded polypropylene tapes for the creation of recyclable all-polypropylene composites. *J Appl Polym Sci* 2007;104(1):118–29.
- [33] Cabrera NO, Reynolds CT, Alcock B, Peijs T. Non-isothermal stamp forming of continuous tape reinforced all-polypropylene composite sheet. *Composites Part A* 2008;39(6):1455–66.
- [34] Alcock B, Cabrera NO, Barkoula N-M, Peijs T. Direct forming of all-polypropylene composites products from fabrics made of co-extruded tapes. *Appl Compos Mater* 2009;16(2):117–34.
- [35] Cabrera NO, Alcock B, Peijs T. Design and manufacture of all-PP sandwich panels based on co-extruded polypropylene tapes. *Composites Part B* 2008;39:1183–95.
- [36] Heijenrath R, Peijs T. Natural-fibre-mat-reinforced thermoplastic composites based on flax fibres and polypropylene. *Adv Compos Lett* 1996;5(3):81–5.
- [37] Stamboulis A, Baillie CA, Garkhail SK, van Melick HG, Peijs T. Environmental durability of flax fibres and their composites based on polypropylene matrix. *Appl Compos Mater* 2000;7(5–6):273–94.
- [38] Peijs T. Natural fiber based composites. *Mater Technol* 2000;15(4):281–5.
- [39] Garkhail SK, Heijenrath RWH, Peijs T. Mechanical properties of natural-fibre-mat-reinforced thermoplastics based on flax fibres and polypropylene. *Appl Compos Mater* 2000;7:351–72.
- [40] Stamboulis A, Baillie CA, Peijs T. Effects of environmental conditions on mechanical and physical properties of flax fibers. *Composites Part A: Appl Sci Manuf* 2001;32(8):1105–15.
- [41] Alcock B, Cabrera NO, Barkoula N-M, Peijs T. Low velocity impact performance of recyclable all-polypropylene composites. *Compos Sci Technol* 2006;66(11–12):1724–37.
- [42] Schimanski T, Loos J, Peijs T, Alcock B, Lemstra PJ. On the overdraw of melt-spun isotactic polypropylene tapes. *J Appl Polym Sci* 2007;103(5):2920–31.

- [43] Alcock B, Cabrera NO, Barkoula N-M, Peijs T. The effect of processing conditions on the mechanical properties and thermal stability of highly oriented PP tapes. *Eur Polym J* 2009;45(10):2878–94.
- [44] Springer GS, editor. *Environmental effects on composite materials*; 1981.
- [45] Schrader EL. Remediation of floating, open water oil-spills-comparative efficacy of commercially available polypropylene sorbent booms. *Environmental Geology and Water Sciences* 1991;17(2):157–66.
- [46] Choi H. Needle-punched cotton nonwovens and other natural fibers as oil cleanup sorbents. *J Environ Sci Health, Part A: Toxic/Hazard Subst Environ Eng* 1996;31(6):1441–57.
- [47] Smith LV, Weitsman YJ. Sea water effects on the fatigue response of polymeric composites. *Prog Durability Anal Compos Sys* 1996:217–23.
- [48] Andreopoulos AG, Tarantili PA. Water sorption characteristics of epoxy resin-UHMPE fibers composites. *J Appl Polym Sci* 1998;70(4):747–55.
- [49] Ellyin F, Maser R. Environmental effects on the mechanical properties of glass-fiber epoxy composite tubular specimens. *Compos Sci Technol* 2004;64(12):1863–74.
- [50] Schimanski T. *High-performance polypropylene structures for eco-friendly, fully recyclable composites*, Netherlands: Technische Universiteit Eindhoven. Ph.D. Thesis; 2002.