



Cyclic pressure on compression-moulded bioresorbable phosphate glass fibre reinforced composites



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ABSTRACT

The use of thermoplastic composites based on poly(lactic) acid and phosphate glass fibres over metallic alloys for clinical restorative treatment is highly beneficial due to their biocompatibility and biodegradability. However, difficulties in achieving a thorough melt impregnation at high fibre contents while limiting polymer degradation is one of the main issues encountered during their manufacture. This paper reports for the first time on the effects of pressure cycling on the mechanical properties of compression moulded polylactic acid-phosphate glass fibre composites. The strength of the composites consolidated under pressure cycling were at least 30% higher than those in which conventional static pressure was used. The marked disparity was attributed to the influence of pressure cycling on the fibre preform permeability, the melt viscosity and the capillary pressure, leading to improved fibre wet-out with respect to static pressure. Implementation of a cyclic pressure appeared to promote the occurrence of transcrystallinity in the polymer matrix as suggested by DSC traces. The fibre content influenced PLA thermal degradation since the matrix molecular weight decreased as the fibre content increased on account of the moisture adsorbed by the glass surface. However, this extent of degradation did not impair the matrix mechanical performance in the composites.

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1. Introduction

Bone atrophy due to stress shielding associated with metallic fixation devices can result in secondary surgery for removal of these implants to prevent long term complications. As such, current research efforts have been focused on the development of novel fully bioresorbable devices capable of eliciting beneficial host responses post-implantation [1–3].

Long-fibre composites fabricated from biodegradable materials, such as polylactic acid 'PLA' and phosphate-based glass fibres 'PGF' can be engineered to initially match (and even surpass) bone mechanical properties and gradually transfer the load to the healing tissue upon degradation. Furthermore, the controlled release of phosphate glasses degradation by-products could stimulate the fracture healing mechanism following *in situ* degradation [4,5].

Amongst the factors that regulate the loading response of composite materials, the fibre content and the adhesion between the matrix and the reinforcement play a critical role [6,7]. From Darcy's law [7], it follows that the higher the viscosity of the polymer melt and the thicker the porous medium, i.e. the higher the fibre volume fraction, the more difficult would be for a given fluid to fully percolate through the interstices of the fibre network.

Intimate interfacial contact is vital not only to fully exploit the strength and stiffness of the reinforcement through effective stress transfer, but also to minimise void content. Voids are detrimental to the composite mechanical properties, due to their action as stress risers and the reduction of the effective matrix-fibre contact area [8]. Moreover, their presence expedites water diffusion, accelerating the loss of mechanical properties in aqueous environments [9].

As a consequence of the high melt viscosity of PLA (and with thermoplastics in general, 100–1000 Pa s [10]), poor quality composites often result from laminates with high fibre to matrix ratio, ultimately hindering the orthopaedic usage of fibre reinforced bioresorbable composites. Conventional approaches utilised in the composite industry to tackle this issue have been: i) the adoption of higher temperatures to reduce the matrix viscosity (enhancing flowability) and ii) increases in the product residence time [11]. Since polylactic acid is a thermally labile thermoplastic which can undergo a rapid loss of molecular weight at temperatures higher than the melting point [12,13], increases in the processing temperature and/or protracted moulding cycles are detrimental to the composite performance.

Alternatively, the consolidation pressure during part forming could be increased to aid in the polymer melt infiltration through the fibre network. However, the fundamental drawback with the increase of pressure is the further compaction of the fibre array, which compromises both the fibre bed permeability and the evacuation of entrapped

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air [7,14]. Therefore, statically applied pressure only has had a limited success in the efficient production of void-free high quality parts with consistent performance under loads (especially at high fibre contents).

In this study it was hypothesised that the implementation of an active pressure profile during part consolidation could address the difficulties imposed by the high polymer viscosity in an extensively used manufacturing technology as compression moulding. This paper reports on the effects of a dynamic compression moulding variant for the production of high volume fraction laminate bioresorbable composites aiming to: i) limit the PLA matrix thermal degradation, ii) minimise void content and iii) significantly improve composite mechanical properties through the optimisation of the pressure scheme applied.

2. Materials and methods

2.1. Melt-drawn fibre spinning and fibre preform production

Phosphate glass fibres of the composition $45\text{P}_2\text{O}_5\text{-}16\text{CaO-}13\text{Na}_2\text{O-}24\text{MgO-}2\text{Fe}_2\text{O}_3$ mol.% and a diameter of $20 \pm 2 \mu\text{m}$ were continuously drawn at 30 ms^{-1} directly from a glass melt using an in-house melt-drawing facility. The parent glass was synthesised from reagent grade NaH_2PO_4 , CaHPO_4 , $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$, P_2O_5 and $\text{MgHPO}_4 \cdot 3\text{H}_2\text{O}$ (Sigma Aldrich, UK) powder precursors which were mixed into a Pt/5% Au crucible (Birmingham Metal Co., UK), and then placed in a furnace for 30 min at $350 \text{ }^\circ\text{C}$ to allow the powders to dry. The crucible was then transferred to a furnace preheated at $1150 \text{ }^\circ\text{C}$ for melting. A 90 min period was allowed for melt homogenisation. Finally the melt was poured onto a steel plate and left to cool to room temperature.

Non-woven unidirectional 'UD' plies were manufactured by drawing fibre onto a traversing drum. The fibres were then sprayed to prevent fibre misalignment using a 5 g/100 mL PLA-Chloroform solution (Resin Ingeo 3251-D NatureWorks®). Following a 2 h period for the complete evaporation of the chloroform, the ply was cut into 121 cm^2 square pieces for subsequent processing. The mass of the plies used for the fabrication of composites with different fibre volume fractions are listed in Table 1.

To be able to assess the degree of impregnation achieved by the respective manufacturing technique, the composites were fabricated from progressively thicker plies as detailed in Table 1.

2.2. Single fibre tensile test

The tensile properties of 60 fibres with a gauge length of 25 mm were evaluated through uniaxial tensile test of individual filaments using a LEX810 Tensile Tester according to standard BS ISO11566. The fibres were randomly selected from a freshly drawn bundle. Prior to testing, the fibre diameter was determined with a Mitutoyo Series 544 LSM-500S Laser diameter gauge.

Evaluation of the tensile strength was conducted using Weibull's statistics assuming a uniform distribution of independent flaws. Weibull parameters were calculated using Minitab® 15 (version 3.2.1).

2.3. Fibre density measurements

Glass fibre density measurements were obtained via use of a AccuPycTM 1330 Pycnometer with a precision of 0.1% calibrated using

a standard steel sphere. Fibre density variability was $\pm 0.02 \text{ g cm}^{-3}$. The analysis was repeated 10 times and the mean value was used for the relevant calculations.

2.4. Composite manufacturing

PLA films of ca. 0.3 mm in thickness were fabricated by pressing 4.5 g batches of PLA resin pellets (Resin Ingeo 3251-D NatureWorks®). The PLA pellets were dried in a vacuum oven at $50 \text{ }^\circ\text{C}$ for at least 24 h prior use and were then placed between two PTFE covered aluminium plates. The plates were then transferred a hydraulic press preheated to $185 \text{ }^\circ\text{C}$. The pellets were held at this temperature for 5 min before compressing at 3 bar for 10 s. Finally the plates were then moved to a second press set at the same pressure for cooling. The PLA films were then trimmed, bagged and stored in a vacuum oven for post-composites at different stages during the manufacturing processes and fibre content processing.

Two series of unidirectional composites of four nominal volume fractions v_f (see Table 1) were manufactured by alternately stacking layers of PGF plies and PLA films inside a purpose-built compression moulding tool with a $4.5 \text{ mm} \times 110 \text{ mm} \times 110 \text{ mm}$ (thickness \times width \times length) cavity. The mould was placed in a hydraulic press at $180 \text{ }^\circ\text{C}$ for 10 min as shown in Fig. 1. Immediately after, the pressure was alternated between 40 and 0 bar for 1 min to assist the evacuation of air trapped during the lay-up, followed by consolidation for 9 min according to two different pressure schemes illustrated in Fig. 1: a) static pressure 'SP' (pressure held constant at 40 bar throughout the 9 min) and, b) cyclic pressure 'CP' (pressure cycled for 1.5 min towards the end of the consolidation stage). Finally the system was cooled under constant pressure to prevent cavitation from crystallisation or thermal shrinkage of the polymer melt.

For each nominal fibre volume fraction in the respective series, at least two composites were fabricated. In addition, pure PLA plates were fabricated from film laminates as control samples.

2.5. 3-Point bending test

Samples of $4.5 \text{ mm} \times 15 \text{ mm} \times 90 \text{ mm}$ (thickness \times width \times length) PLA and composite plates were characterised through 3-point bending test in accordance with BS EN ISO 14125:1998 standard. Three samples for each composite fabricated were tested using a Hounsfield Series S testing machine with a cross-head speed of 1 mm min^{-1} , and a 5 kN load cell.

2.6. Scanning Electron Microscopy (SEM)

The 3-point bending test coupons were mounted in epoxy resin, polished and sputter coated with platinum and examined using a Philips XL-30 scanning electron microscope with an accelerating voltage of 10 kV in secondary electron mode.

2.7. Differential Scanning Calorimetry (DSC)

The thermal properties of the composite's polymer matrix were determined with a DSC Q10 (TA Instrument, USA). Prior to use the equipment was calibrated with mercury and indium standards. Thin composite slices, cut in the transverse direction using a diamond saw, were loaded into sealed aluminium pans and heated under nitrogen flow of $50 \text{ cm}^3 \text{ min}^{-1}$ from $30 \text{ }^\circ\text{C}$ to $190 \text{ }^\circ\text{C}$ at a rate of $5 \text{ }^\circ\text{C min}^{-1}$. A 5 min isothermal hold was allowed before cooling at a rate of $20 \text{ }^\circ\text{C min}^{-1}$ back to $30 \text{ }^\circ\text{C}$. The equipment was allowed to equilibrate at this temperature before resuming a subsequent run with the same parameters as those previously described.

On account of the fibre content, the mass of the composite slices was adjusted according to their experimental volume fraction to keep the polymer matrix mass constant at ca. 13 mg. The samples were dried under reduced pressure for 48 h (0.01 mbar) at $30 \text{ }^\circ\text{C}$ and stored in a

Table 1
Composition of the laminates assembly to achieve the target volume fractions.

Sample codes	v_f	PLA mass (g)	PGF mass (g)	Plye mass (g)	No. of plies
PLA-plate	0	67.31	–	–	–
Vf-15	0.15	57.21	21.35	1.33	16
Vf-25	0.25	50.48	35.58	2.54	14
Vf-35	0.35	43.75	49.81	4.15	12
Vf-45	0.45	37.02	64.04	6.40	10

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