3rd International Conference on Natural Fibers: Advanced Materials for a Greener World, ICNF 2017, 21-23 June 2017, Braga, Portugal

Effect of time at temperature for natural fibres

Alexander Hart[a] and John Summerscales[a]

[a]Advanced Composites Manufacturing Centre, MAterials and STructures (MAST) research group, School of Engineering, University of Plymouth, Plymouth, PL4 8AA, United Kingdom.

Abstract

The manufacture of natural fibre reinforced polymer matrix composites is generally assumed to be limited to temperatures not exceeding 200°C. However, given the covalent bonding in the various constituents of natural fibres, they should retain properties to an extent dependent on the time at temperature. This study considers fibre mechanical properties after heating in air for various temperatures and times.

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Peer-review under responsibility of the scientific committee of the 3rd International Conference on Natural Fibers: Advanced Materials for a Greener World.

Keywords: Jute; Natural fibres;Time-at-temperature

Nomenclature

RH   relative humidity
T_d Degradation temperature
T_g Glass transition temperature
T_m Crystalline melting point

* Corresponding author. Tel.: +44-1752-5-86150 ; fax: +44-1752-5-86101.
    E-mail address: jsummerscales@plymouth.ac.uk

1. Introduction

There is increasing interest in the use of natural fibres as the reinforcement for polymer matrix composites. Natural fibres may come from animal, plant or mineral sources, but the current principal interest is for bast (stem) fibres from plants [1-5]. Plant materials primarily contain cellulose fibres in a matrix of hemi-celluloses and pectins with increasing proportions of lignins arising as the plant ages.

Cellulose is a hydrophilic molecule, so it is essential for composite manufacturing processes that the reinforcement fibres is dried before use to prevent trapped moisture becoming voids in the composite matrix. ISO12215-4:2002 [6] requires that "The relative humidity in the moulding shop shall be maintained within the limit recommended by the material manufacturers". Armstrong and Barrett [7] recommend drying cycles (for aramid honeycomb) of 16 h at 40°C, 3 h at 100°C or 1 h at 160°C, with the materials then used within 1 h of drying. Davies [8] suggested that dried cores should be stored in a dedicated room at 50°C and <10% RH.

Dry cellulose is variously reported to have glass transition temperatures (T_g) in the range 84-250°C with T_g decreasing with increasing water content. Roig et al report that the T_g of cellulose is strongly dependent on the degree of crystallinity [9]. Szcześniak et al. reported colour changes at 180°C and significant weight losses (~70%) between 220°C and 300°C [10]. Cellulose normally decomposes in air before melting because of the extensive hydrogen bonding between the cellulose molecules. The melting temperature of cellulose is reportedly in the range 500-518°C (presumably under inert gas or in vacuo) [11].

Ligno-cellulosic fibres degrade at around 200°C in air [12-14], so there are few commercial thermoplastic matrix systems available for load-bearing applications at ambient temperature (i.e. below T_g to avoid creep) yet processed without degradation of the fibres ($T_m \approx T_g+200°C$). A notable exception is poly(lactic acid) (PLA) with T_g = 55-65°C, crystalline melting point, T_m = 120-170°C and degradation temperature, T_d, just above 200°C. PLA has a narrow process temperature window. PLA-matrix composites should be able to take stress at ambient temperatures without creep occurring.

Use of other polymer matrix systems might be possible if the time at higher temperatures is minimised (or if processing is under vacuum or in an inert atmosphere, although the process costs will inevitably be higher). This paper seeks to identify a process window for time and temperature where natural fibres can processed without significant degradation of their mechanical performance.

2. Experimental materials and methods

This study used fibres from a well-characterised single batch of quasi-unidirectional jute with an areal weight of 880 gm^{-2} [15-22]. Batches of fibre were heated in an oven to set temperatures (180-220°C step 10°C) for predetermined times (5, 10, 15, 30, 60 and 120 minutes). The individual fibres were mounted using epoxy resin on cards, based on the Grafil Test 101.13 for carbon fibres, and tested for apparent* filament tensile modulus and strength with a 50 mm gauge length. Five samples at each combination of temperature and time were tested and analysed. A number of filaments failed before they were ready for test and are not included in the analysis. Testing was undertaken in compliance with BS ISO 11566:1996 on an Instron single column test machine (serial number 3345K1669 calibration certificate E23032116110441 expiring March 2017, i.e. during the test series) with an Instron 10 N force transducer (model 2519-101, serial number 64650), with at 0.5 mm/min under ambient conditions. Fibre diameters were measured after failure and the analysis assumed circular cross section for the fibres. For true moduli and strength, the values should be increased using the fibre area correction factor* for (this batch of) jute fibres at 1.42 as established by Virk et al [20].

3. Results

Figure 1 shows the change in colour with increasing time and increasing temperature. Figure 2 shows the Young's moduli of the jute fibres plotted against "apparent" jute fibre diameters. Figure 3 presents the mean tensile moduli for the fibres plotted against time at temperature. Figure 4 separates the moduli for fibres heated at low and high temperatures respectively against heating time. Figure 5 shows the mean strain to failure for all fibres plotted against time at temperature.
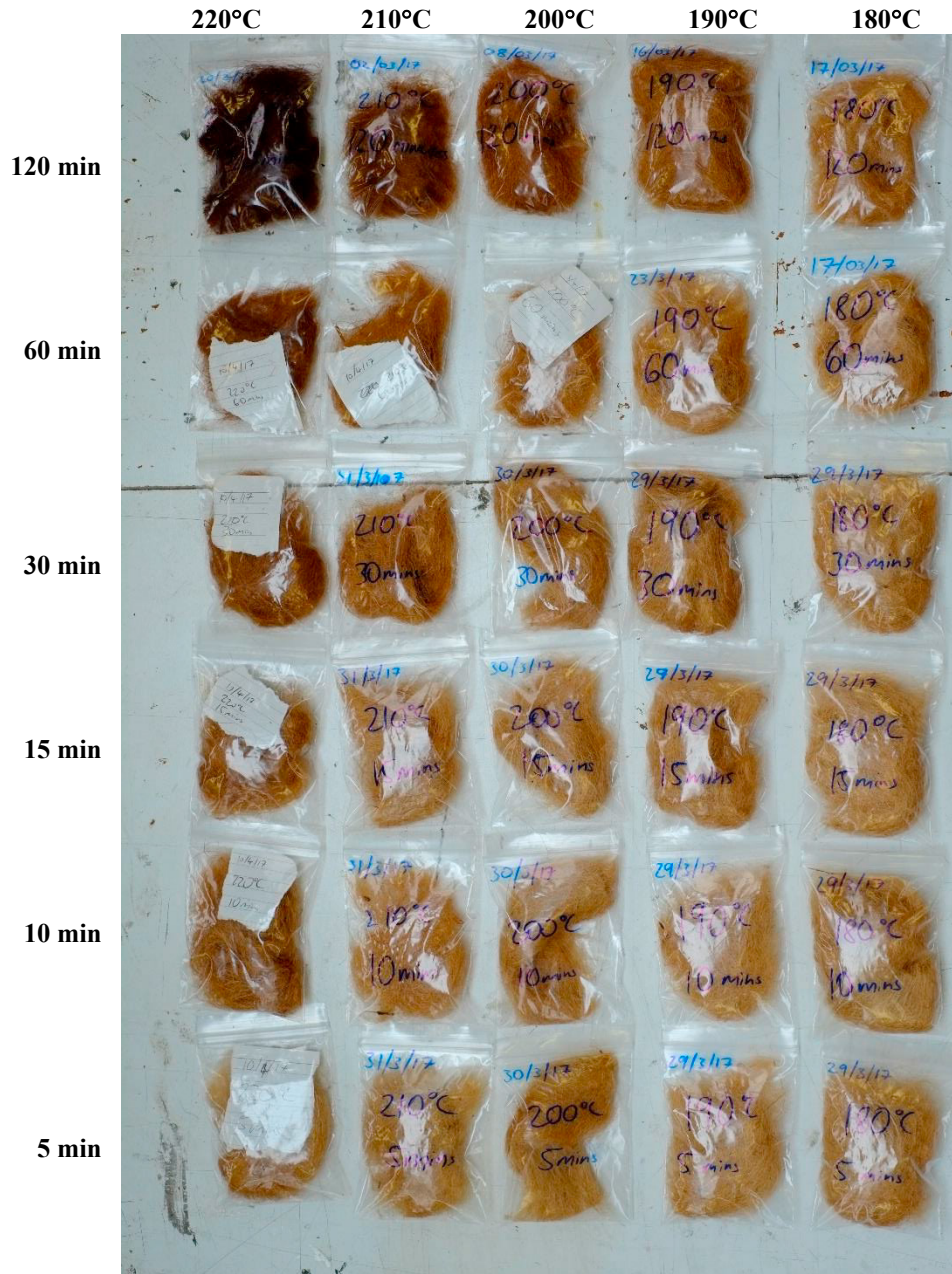

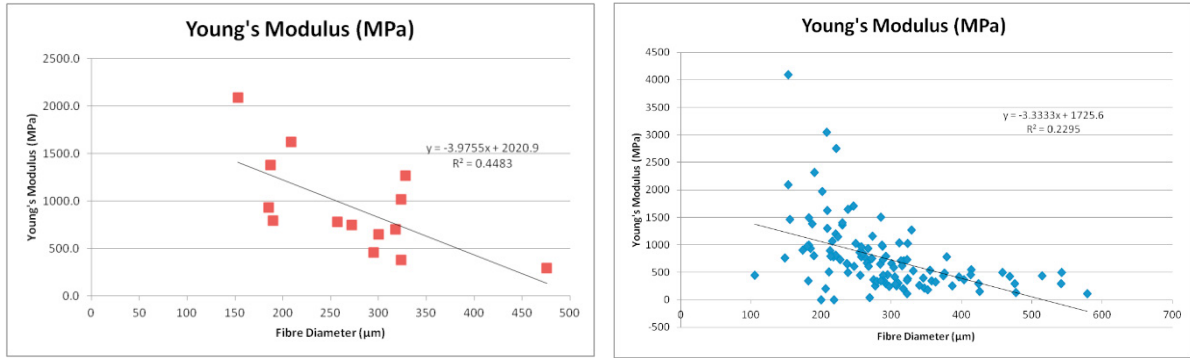Figure 1: Fibre discolouration with time and temperature

Figure 2: Young's modulus of the jute fibres plotted against "apparent" jute fibre diameters: fibres heated at 220°C between 5-15 minutes (left), all fibres (right)
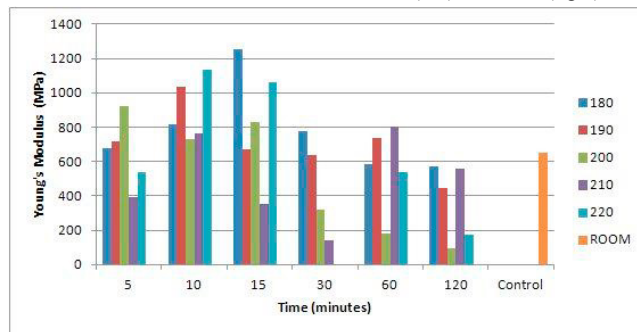

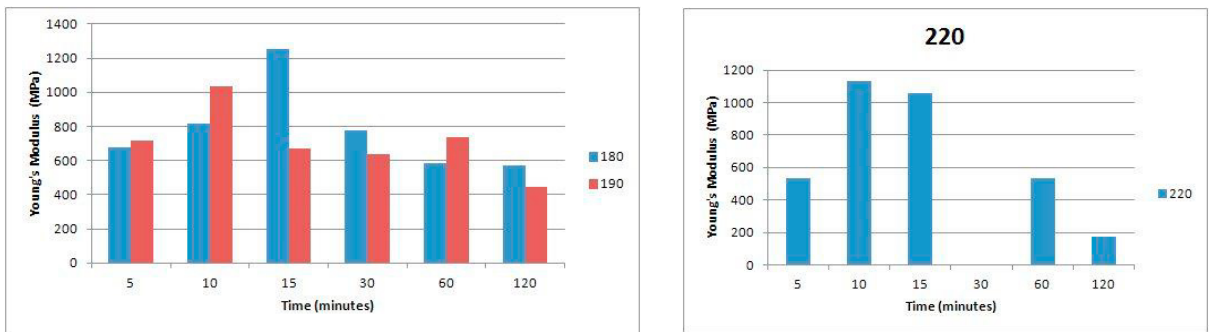Figure 3: Mean Young's moduli for all fibres plotted against time at temperature

Figure 4: Young's moduli for fibres heated at 180°C or 190°C (left) or 220°C (right) plotted against heating time.
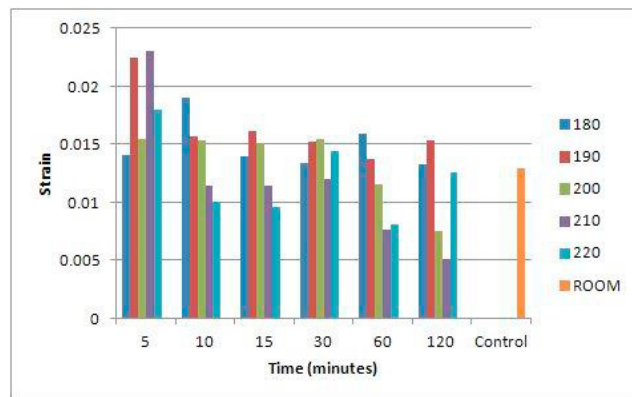

Figure 5: Mean strain to failure for all fibres plotted against time at temperature

4. Discussion

The tested fibres had an average "apparent" diameter in the range 100-600 μm (Fig.1). The fibres are thus considered to be technical fibres rather than ultimate fibres. The decrease in tensile modulus with increasing fibre "apparent diameter" has been reported for jute [23], flax [24] and nettle [25] fibres. This factor will influence the following discussion: in retrospect, a greater number of samples should be tested at each condition to reduce variation in the data and increase the validity of the research results.

As a broad conclusion, the data obtained suggests that at temperatures below 200°C, there is no significant change in properties with heating times up to 30 minutes, but extended heating may compromise the mechanical properties. There appears to be an increase in properties for fibres heated for short periods. However, the recorded time may over-estimate the heat input as opening the oven door to load or remove samples caused drop in temperature. The oven consistently took around four minutes to return the set temperature.

At higher temperatures, the loss of properties appears to begin at shorter heating times. For samples heated between 15-120 minutes, there is a clear decline in the Young's moduli with increasing temperature.

The tests were conducted in the context of an undergraduate project with a nominal 50 days allowed for the entire project. In consequence, time constraints meant that it was only possible to obtain valid results for five samples at each combination of temperature and time. The experiments undertaken should be regarded as preliminary results, with a number of limitations now recognised:
- The recorded temperatures in the oven were measured using a thermocouple remote from the fibres,
- Fibres were assumed to be perfectly aligned with the test machine axis,
- Fibre "apparent diameters" were measured with high accuracy, but average values for each fibre were calculated to three significant figures for calculation of mechanical properties. These data do not acknowledge the variation in the "apparent diameter" which is inherent in natural fibres.
- Some "toe-in" on the load/deflection curves may influence the determination of the modulus from the reported data,
- The fibres from the 30 minute exposure at 220°C separated from the test card on failure and hence were not available for "apparent diameter" measurement.

In respect of the manufacture of natural fibre composites, the smallest reduction in performance should be achieved by processing at the lowest possible temperature for the shortest time. However, as a rule-of-thumb, the viscosity of thermosetting resin decreases by one-half for a 10°C rise in temperature, so process times will be optimal for a specific combination of temperature and duration. Processing under vacuum or inert gas (e.g. N_2 or CO_2) may delay degradation, but will inevitably increase process costs.

5. Conclusions

Following heating at set temperatures (180-220°C step 10°C) for predetermined times (5, 10, 15, 30, 60 and 120 minutes), the mechanical properties of jute fibres were measured under ambient conditions. The results indicate that degradation occurs more rapidly at the higher temperature and that extended heating at the lower temperatures compromises properties. The data available is from a limited set of tests and hence it has not yet been possible to determine optimum processing temperature-time values.

Acknowledgements

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