Solid epoxy resin systems for automated composite manufacturing

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ABSTRACT

Thermosetting resins were formulated by mixing solid epoxy components, hardener and accelerator at elevated temperature. These mixtures were ground into uncured, solid resin powders and dispensed onto carbon fibres using a powder printer prior to vacuum curing. Using accelerators reduced curing time from 8 h to 16 min. Manufactured cross-ply carbon fibre laminates (0°/90°/0°) had comparable mechanical properties to composites manufactured using a liquid resin system. Tack behaviour of solid epoxy resin formulations was investigated and results compared to a conventional liquid resin system. Master curves of the storage and loss moduli of both solid and liquid resin formulations were generated by applying time-temperature superposition to shear moduli between 1 and 100 rad/s at multiple temperatures. This approach was used to determine optimum processing parameters to enable resin tack for automated fibre placement, demonstrating the importance of temperature control with respect to deposition rates.

1. Introduction

Carbon fibre composites could potentially be used in a wider range of sectors if significant cost savings can be made [1–3], for example by using less expensive precursors to manufacture carbon fibres [4,5] or utilising recycled materials [6–8]. On the other hand, using inexpensive matrix formulations and producing intermediates (alternatives to prepregs) can also reduce costs. The use of low cost feedstock materials in technologies such as automated tape laying (ATL) and automated fibre placement (AFP) [9–11], which are typically limited to the aerospace industry, could significantly broaden the application areas of automated manufacturing of composites for high volume production. These technologies use wide (ATL) and narrow (AFP) prepreg tapes for material lay-down. ATL is only suitable for large, gently contoured parts, such as wing skins or fuselage sections [12–16], whereas AFP is preferable for more complex part geometries [9]. Aerospace roadmapping [17] has identified AFP as a high priority research area to target deposition rates of around 100 kg/h to increase productivity. However, deposition rates of only 29.2 kg/h for ATL and 41.3 kg/h for AFP [18] were reported by

Lukaszewicz et al. [18–20]. Deposition rates are generally limited by part geometry, material formats and the dynamic response of large machines for both technologies [18,20,21]. Automated dry fibre placement (ADFP) has recently been investigated to potentially increase preforming rates and reduce material costs by using dry slit tapes [22–24] rather than prepregs. However, ADFP preforms suitable for liquid composite moulding require heavily processed dry fibre tapes, resulting in high intermediate material costs [25,26]. They also suffer from much lower tack than prepreg tapes, significantly limiting production rates.

Tack, the pressure sensitive adhesion of uncured resins, is considered to be important for the robustness of automated placement techniques [27] and could be the key to unlocking higher deposition rates and preform quality. Sufficient tack levels are required to deposit tapes either onto the mould substrate or to previously deposited plies in the preform. Typically AFP or ATL processes use a constant but elevated temperature [28] to achieve resin tack within a suitable operating window. However, this temperature is usually determined by trial and error and it is difficult to optimise the tack force with changes in

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deposition rate. Deposition temperature may be controlled dynamically, by adjusting the temperature to be proportional to the rate, but this relies on accurately modelling the heating power output to control the nip-point temperature [22]. Crossley et al. [29] measured the tack-rate response for a range of prepreg materials and showed that the processing temperature range depends on the deposition rate. Material tack is experimentally evaluated using standard test frames with limited strain rates (<500 mm/min). Therefore, to determine tack at higher deposition rates (>500 mm/s) the relationship between temperature and rate must be extrapolated to relevant production rates. This is possible using the time-temperature superposition (TTS) principle, applying a shift factor similar to that used in the empirical Williams-Landel-Ferry method [4,30].

In this paper, we describe a low cost solid epoxy resin formulation and a composite manufacturing method utilising this solid epoxy resin. We will show that our solid resin formulation is suitable to serve as composite matrix. Therefore, cross-ply composites are prepared using a resin powder printing head and their flexural properties determined. By using the powder printing head, we investigate the potential of directly dispersing solid epoxy resin powders onto dry fibre tapes. This could open up the possibility to develop a low-cost ADFP method by directly depositing dry carbon tows onto resin powders, which were dispersed beforehand using a powder printing head, rather than using more expensive prepreg or dry fibre slit tapes with a thermoplastic veil or binder. The other main advantage is that the resin flow distance is significantly reduced compared to liquid resin infusion processes, reducing the likelihood of dry fibre spots [31,32]. Furthermore, the tack-rate response of our solid epoxy resin formulation is compared to an equivalent deposition onto a film of a liquid epoxy resin formulation, typically used in prepregging.

2. Experimental methods

2.1. Materials

Solid epoxy resin (Araldite GT7071) and powdered accelerator (Dyhard UR500) were kindly supplied by Huntsman (Duxford, UK). A powdered dicyandiamide hardener (Dyhard 100S) was supplied by AlzChem (Trostberg, Germany). The flexural performance of composite specimens produced by the solid epoxy system was compared against specimens produced by a conventional liquid epoxy. A liquid epoxy system provided by Huntsman (Duxford, UK) (XB 6469 and Aradur 2954) was used to produce comparison plaques for flexural testing.

The tack performance of the solid epoxy system was compared against a second liquid epoxy resin (Huntsman Araldite LY1556, Aradur 1571, Accelerator 1573 and Hardener XB 3403), which was selected because it is suitable for B-stage curing.

Unidirectional (UD) non-crimp fabric (consisting of Toray T700SC-50C 12k carbon fibres), kindly provided by Hexcel (Leicester, UK), was used to produce composite samples.

EpoxiCure 2 resin and hardener, purchased from Buehler (Düsseldorf, Germany), was used for the preparation of composite microsections.

2.2. Formulating solid epoxy resins

Solid epoxy granules and hardener powder (4 wt%) were weighed on a balance and then mixed in a sealed container at room temperature. The resulting mixture was blended at 80 °C for 5 min in a kneader (Plastograph, Brabender, Duisburg, Germany, equipped with a 3 zone W50EHT mixing unit) at mixing speeds ranging from 5 to 100 rpm. Resins containing 1 or 2 wt% accelerator (all with 4 wt% hardener) were processed at a mixing temperature of 65 °C to prevent premature curing. The resin formulations and their properties are listed in Table 1.

The resin mixtures were collected, allowed to cool and then ground into powders using a mechanical grinder (Karcher UM 620). The powders were sieved (mesh 40; aperture size 425 μm, Retsch) to obtain a uniform particle size of approximately 25 μm. The solid resin powders were stored at 5 °C to prevent any unwanted curing.

2.3. DSC analysis of solid epoxy resins

The thermal properties of the uncured and cured resins were analysed using differential scanning calorimetry (DSC) (Discovery DSC, TA Instruments, Eschborn, Germany). Samples were heated and cooled at a rate of 10 °C/min in a nitrogen atmosphere. All specimens were analysed from 0 °C to 300 °C, cooled back to 0 °C and then re-heated to 300 °C.

2.4. Manufacturing carbon fibre reinforced composites

Release agent (Frekote 700-NC, Henkel, Germany) was applied to the surface of a hotplate (HP 1836-AH, Wenesco Inc., USA) in an area outlined by masking tape. Once the release agent had dried, a layer of polyimide release film (Upilex-25S, UBE, Osaka, Japan) was placed directly onto it and the masking tape removed. Vacuum bag sealant tape (Airdam 1, Airtech, Luxembourg) was placed around the edge of the release agent coated area (Fig. 1c). Three 80 mm × 80 mm carbon fibre plies were prepared along with four batches of resin powder, each weighing 1 g. The resin powder was dispersed on the carbon fibre layers using a vibrating nozzle dispenser (ESI Fig. S1 & S2, Fig. 1a) covered with a mesh (mesh 30; aperture size 500 μm), developed by us. This nozzle was attached to a universal CNC model-making machine (Stepcraft 420, Stepcraft GmbH, Iserlohn, Germany). The first carbon fibre layer was evenly coated with 1 g of resin powder on both sides (corresponding to an aerial weight of 156 g/m^2) and placed on top of the release film (Fig. 1b). The next layers of carbon fibre were powder coated on one side only and were subsequently placed on top of each other (fibre orientation 0°/90°/0°). The three carbon fibre layers coated with solid resin powder were surrounded by flow media (70 × 40 mm, Composite Envisions, Airtech Greenflow 75 Resin Flow Medium; Fig. 1c) to aid air removal. Release film was placed on top of the sample, followed by vacuum bagging film. A through-bag connector with a ¼turn valve and barbed hose fitting was placed at one end of the bag away from the sample, which was used to connect the vacuum bag to a vacuum pump. A metal plate (500 g) was placed on top of the sample to keep the stacked coated carbon fibre layers in place. The specimens coated with resin containing 1 wt% accelerator were heated from 37 °C to 65 °C and held at this temperature for 10 min in order to melt the solid

Table 1
Solid epoxy resin compositions and properties.

Accelerator content [wt.%]	Hardener content [wt.%]	Uncured T$_g$[†] [°C]	Cured T$_g$[†] [°C]	Normalized curing enthalpy [J/g]	Onset of curing [°C]	Cured T$_g$[††] [°C]
0	4	35*	114 ± 2	187	180	107*
1	4	35*	108 ± 5	215	126	111 ± 3
2	4	35 ± 1	108	189	121	113 ± 3

[†] Determined using DSC.

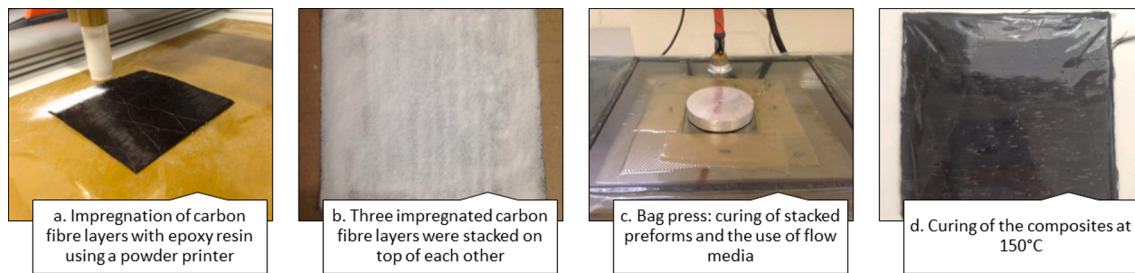[††] Determined by DMTA in bending.

* Standard derivation below 0.5.

Fig. 1. Manufacturing process of carbon fibre reinforced composites. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

resin completely and to degas the stack prior to curing. The temperature was then increased to 85 °C with vacuum applied for another 10 min and this procedure was repeated at 125 °C. Finally, the temperature was increased to 150 °C to cure the composites. The vacuum was switched off upon reaching 150 °C. Composite preforms impregnated with matrix containing 1 wt% accelerator were cured at 150 °C for 16 min. Composites without accelerator were heated from 47 °C to 140 °C and held at 140 °C for 10 min to degas. The temperature was then increased to 150 °C and held for 8 h. The vacuum was shut off when 150 °C was reached. All composites were then slowly cooled down to room temperature by switching off the hotplate, which took approximately 2 h. The void content of the composites was determined in accordance to ASTM D2734-94. Acid digestion was performed following procedure B detailed in ASTM D3171-99.

2.5. Flexural testing of composites

The composites were tested in three-point bending using a universal test frame (Instron 5969) equipped with a 50 kN load cell. The tests, in accordance with ASTM D7264-07, were performed using a crosshead speed of 1 mm/min on specimens with a 32:1 span-to-thickness ratio. The diameter of the loading nose and supports was 6 mm. All tests were performed at room temperature. The flexural modulus E_f of the specimens was calculated using Eq. (1):

$$E_f = \frac{L^3 m}{4bh^3} \tag{1}$$

where m is the gradient of the linear section of the load-displacement curve, L the support span, b the beam width and h the beam thickness. The flexural strength of the composites σ corresponding to the maximum stress at the outer surface at the mid-span of the specimen was calculated using Eq. (2):

$$\sigma = \frac{3PL}{2bh^2} \tag{2}$$

where P is the applied force at failure. Values from at least nine specimens were averaged and the standard deviation reported.

2.6. Preparation of composite microsections

Composite microsections were prepared in order to analyse cross-sections of the composites. Potting resin (EpoxiCure 2, Buehler, Germany) was mixed, poured over the composite samples and then cured overnight at room temperature. The samples were polished using an EcoMet 250/AutoMet 250 (Buehler, Germany). Four sandpapers with varying grit sizes were initially used (SiC P180, P320, P600 and P1200), followed by 3 diamond dispersions (9 μm, 3 μm and 1 μm). The polished samples were then examined using an optical microscope (Axioplan, Zeiss, Germany) equipped with a digital camera (AxioCam MRc5 5-MP colour camera, Zeiss, Germany).

2.7. Resin film formation

Resin films were produced to investigate the potential of using this resin formulation in automated dry fibre placement. The solid resin (and the four-component liquid formulation used for control specimens) was deposited onto 3 mm thick stainless-steel plates (75 mm × 135 mm). The solid and liquid resin formulations were dispensed in a continuous line profile with a width of 70 mm and a step height of 5 mm for the solid resin and 50 mm for the liquid resin. To illustrate this profile, a drawing of the deposition pattern on a tack testing plate is shown in ESI Fig. S3. The solid resin was deposited using the dispersion nozzle mounted to the Stepcraft printer.

Liquid resin spray equipment was used to create films using the liquid epoxy resin [33,34]. A robot arm was used to direct the atomised spray of the liquid epoxy resin at an angle of 90° to the tool substrate. Following the film deposition, the resin was subjected to a B-stage process at 80 °C for 25 min to prevent dewetting of the resin into droplets on the plate. B-staging advanced the degree of cure to approximately 22%, as determined by DSC. A second stainless steel plate of equal size was placed on top of the wetted surface during B-staging. This was wrapped in release film to prevent adhesion to the second surface. The release film was removed once the viscosity of the resin had sufficiently increased.

2.8. Tack testing

Adhesion of fibres to the resin substrates (tack) was characterised to explore the suitability of our solid resin formulation for automated fibre placement processes. Tack results were compared to those of a conventional formulated (liquid) resin system, typically used for prepreg production [35] and representative of slit tapes. Rate and temperature dependent tack was measured using the testing method and equipment developed by Crossley et al. [29,30]. Tack testing was performed using a universal test frame (Instron 5985) equipped with an environmental chamber and a 1 kN load cell. Tack test specimens with dimensions of 75 mm × 215 mm were produced by placing dry carbon fibre tows onto the resin films prior to tack testing, using an AFP machine at Formax (Leicester, UK). Prepreg backing paper was used to avoid contact between the first 70 mm of fibres and the resin film to prevent initial contact with the compaction roller. The fibre/resin specimens were then placed between the compaction and guide rollers of the tack testing rig (ESI Fig. S4). The excess fabric length was clamped to the load cell fixture and the temperature of the environmental chamber was raised to the required test temperature, allowing the apparatus and sample to equilibrate for 15 min at each test temperature. A compaction force of 100 N was applied to the sample using two tension springs by releasing the spreader bar. Seven solid resin specimens were tested between 60 and 81 °C and nine B-staged liquid resin specimens between 19 and 35 °C using a constant extension rate of 100 mm/min. The fibres and resin are initially separated by the backing paper as the material is drawn through the rig, therefore the force generated is the result of the bending stiffness of the fabric. The fibres are compressed into the resin

film before separating around the compaction roller. The total peel resistance is the sum of the fabric stiffness and the tack force, as shown in Fig. 2. The tack force is obtained by subtracting the fabric bending stiffness from the total peel resistance [29,30].

2.9. Rheological characterisation of resin films

Frequency sweeps were conducted on 570 μm (±30 μm) thick, 25 mm diameter resin film specimens between parallel plates at shear rates of 1 to 100 rad/s to determine the dynamic viscosity, storage and loss moduli (Anton Paar MCR302 equipped with an oven). These sweeps were performed at 10 °C intervals from 20 to 50 °C for the B-staged liquid resin system, significantly below the cure temperature of the resin. A higher temperature range (50–150 °C) was required for the solid resin system without accelerator. This temperature was held for a relatively short period of time in comparison to the cure cycle of 8 h at 150 °C. The temperature was increased by 5 °C/min between frequency sweeps.

2.10. Time-Temperature superposition

To scale the tack testing rates (100 mm/min) to the fibre deposition rate during fibre placement (approximately 500 mm/s), the time-–temperature superposition (TTS) principle was used. This enables the shear moduli of the resin system, determined from the rheological data, to be shifted to generate a master curve for a greater range of test speeds for a given reference temperature T_0. This curve was obtained by optimising the empirical shift constants, C_1 and C_2, within the Williams-Landel-Ferry (WLF) equation [4]:

$$log(\alpha(T)) = \frac{-C_1(T - T_0)}{C_2 + (T - T_0)} \qquad (3)$$

The C_1 and C_2 values are determined by the best-fit between the overlapping (shifted) data to produce the master curve. This is achieved using the open-source optimiser Reptate [5]. The WLF theory was then used to calculate the shift factor $\alpha(T)$ of the storage and loss moduli at all test temperatures to the reference temperature.
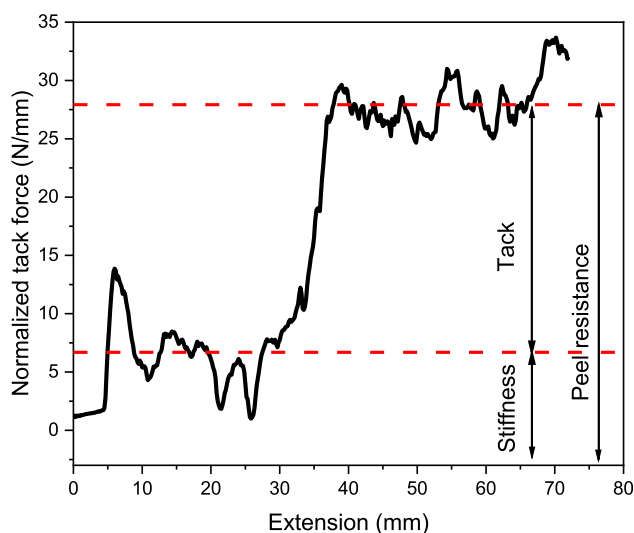
3. Results and discussion

3.1. Solid epoxy resin characterisation

The uncured milled solid epoxy resin without accelerator had a Sauter Mean Diameter (SMD) of 22 μm. The SMD of the uncured resins containing 1 wt% and 2 wt% accelerator was 28 μm and 24 μm, respectively. The addition of accelerator did not affect the ability to process the powder using the dispensing head. As shown in Table 1, the uncured resins containing 1 and 2 wt% accelerator had similar thermal properties to the resin without accelerator. After blending the resin formulations in the kneader at 65 °C, the uncured T_g remained 35 °C for all resin formulations. This indicated that no pre-curing of the resin occurred.

The onset of curing occurred at 180 °C for the resin without accelerator as seen in the first DSC heating curve (Fig. 3a, Table 1). Fully curing the resin for 8 h at 150 °C resulted in a cured T_g of 114 ± 2 °C (Fig. 3b, Table 1). The cured T_g of the solid resin is similar to that of commercially available liquid epoxy resins, such as Araldite® LY 1556 [36]. The onset of curing for 1 and 2 wt% accelerator occurred earlier at 126 °C and 121 °C, respectively. Fully curing the resin resulted in a cured T_g of 108 ± 5 °C for resins containing 1 wt% accelerator. The similar cured T_g for the resins with and without accelerator suggests that the degree of crosslinking was similar. The mechanical T_g (determined
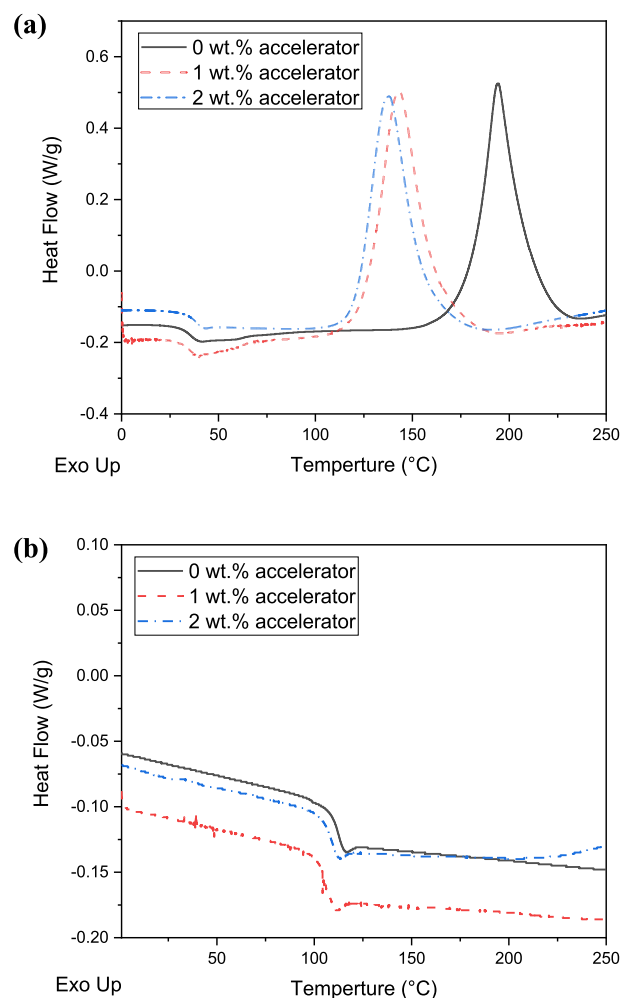
(a)

(b)

Fig. 3. First heating DSC curves of the uncured resin formulations containing 0, 1 and 2 wt% accelerator (a) and of fully cured resins (b). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 2. Example tack force calculation of a solid resin specimen. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

by DMTA) of the resins with and without accelerator was 107 and 111 ± 3 °C, respectively. These results confirm the similar degree of cross-linking of the resins. The resins containing 2 wt% accelerator were not used further due to the higher void content in composites manufactured using this formulation.

3.2. Time-temperature superposition and tack performance

Tack testing was performed at a constant rate (100 mm/min) to measure the tack force for the solid epoxy and B-staged liquid epoxy as function of temperature. Both resin systems exhibited a single tack force peak across the tested temperature range. The peak tack force for the solid resin occurred at approximately 60–85 °C and the peak for the B-staged liquid resin it occurred at 20–35 °C (Fig. 4). If the temperature used during fibre deposition is lower than the peak temperature range, the fibres exhibit immeasurable tack. On the other hand, if the temperature exceeds the peak temperature, the reduction in resin viscosity allows for slippage between the specimen and steel plates, reducing the measurable tack between the two components. A similar peak tack force of 21 N per 75 mm width can be observed for the solid resin at 65 °C and for the liquid resin at 27 °C, indicating that the tack performance is comparable for both systems albeit at different temperatures. To minimise the frequency of defects during AFP processing, high tack is desired to ensure that the tapes adhere to the tool surface or previously deposited plies. Therefore, the peak tack forces indicate the optimal deposition temperature for each epoxy system.

Shift factors were obtained by applying time-temperature superposition [30] to each measured tack value at test temperature T and constant feed rate (100 mm/min) to calculate the shifted rate at a constant reference temperature T_0. These shifted values represent the new rate that exhibits the measured tack force (at 100 mm/min) at the reference temperature. This enables appropriate reference temperatures to be investigated, which would enable peak tack at the target deposition rate of 500 mm/s. This rate is more appropriate for the AFP process, but is beyond the capabilities of the testing machine.

To achieve this, the shear moduli of the resin systems were investigated to determine the shift parameters. The shear moduli, obtained during frequency sweeps, were shifted with respect to angular frequency (ESI Fig. S5) to enable the determination of the empirical constants, C_1 and C_2, and their use in Eq. (3). By calculating the shift factor for each data point, depending on the test temperature, master curves of storage and loss moduli were created for each reference temperature T_0, with

respect to angular frequency (ESI Fig. S6). The constants determined for the solid and liquid B-staged resin systems are summarised in Table 2.

These shift constants derived from the shear moduli are applicable to the tack data, enabling shift factors to be calculated depending on the test temperature of each data point. These shift factors can be used to determine the shifted rate at each of the reference temperatures. This is shown in Fig. 5.

The solid resin has a peak tack force of approximately 21 N per 75 mm width at a temperature of 90 °C, between deposition rates of 100–200 mm/s. However, if the temperature is increased by 20 °C to 110 °C, the tack force is approximately 67% lower at this rate. The peak tack force for the liquid resin was observed at a similar rate of 100–200 mm/s but for a reference temperature of just 40 °C. However, a similar reduction in tack force of the solid resin occurs for the liquid system (at the same rate), when the reference temperature of the liquid resin is increased by only 10 °C. This indicates that the liquid resin is more sensitive to rate and temperature changes than the solid resin system, i.e. the peaks shift further with respects to rate, for similar reference temperatures.

3.3. Flexural properties of cross-ply carbon fibre reinforced composites

The flexural modulus and strength of the composites manufactured using the solid resin system without accelerator were 74 ± 7 GPa and 1127 ± 183 MPa, respectively (Table 3). Specimens manufactured using the solid resin system containing 1 wt% accelerator had a lower flexural modulus (61 ± 9 GPa) and strength (1070 ± 144 MPa). Within the errors, the overall flexural properties of all composites manufactured from the solid resin system (with and without accelerator 1 wt% accelerator) did not change significantly. Composites manufactured using the same fibres but using a liquid resin system (Table 3) had a flexural modulus and strength of 69 ± 5 GPa and 1116 ± 86 MPa, respectively. Please note that the fibre volume fractions of the composites manufactured using solid resins formulations were significantly lower as compared to those produced using the liquid resin, however despite this, all flexural properties are comparable with an error.

3.4. Cross-sections of carbon fibre reinforced composites: Void content and fibre alignment

Cross-sections of cross-ply (0°/90°/0°) composites manufactured using the solid epoxy resin with and without 1 wt% accelerator were analysed to observe voids and fibre alignment. Using flow media during the manufacturing of composites enabled the void content to be significantly reduced from 8% to less than 2%. Fig. 6a shows a micrograph of a typical composite cross-section produced using the resin without accelerator.

Cross-ply composites produced using solid resin formulations, with and without accelerator, were well impregnated (Fig. 6). Whilst the resin containing accelerator gelled faster, the void content remained low at 2.0 ± 0.7%, compared to 1.8 ± 0.5% for the baseline composite produced using the resin without accelerator. The carbon fibres are well distributed within the matrix, with no significant difference in fibre alignment observed. The addition of the accelerator had no significant effect on the void distribution as shown in Fig. 6.
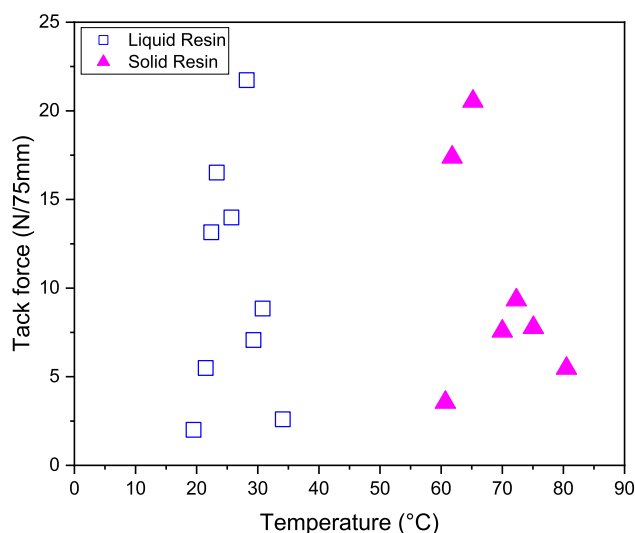

Fig. 4. Tack force for liquid and solid resin systems at a constant rate 100 mm/min. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 2
Optimum parameters C_1 and C_2, of the WLF equation at their reference temperatures.

Solid Resin				Liquid Resin			
Reference temperature [°C]	70	90	110	Reference temperature [°C]	30	40	50
C1	6.2	4.8	3.9	C1	6.6	5.6	4.8
C2	69.5	89.7	109.4	C2	52.8	62.8	72.8

(a)

(b)
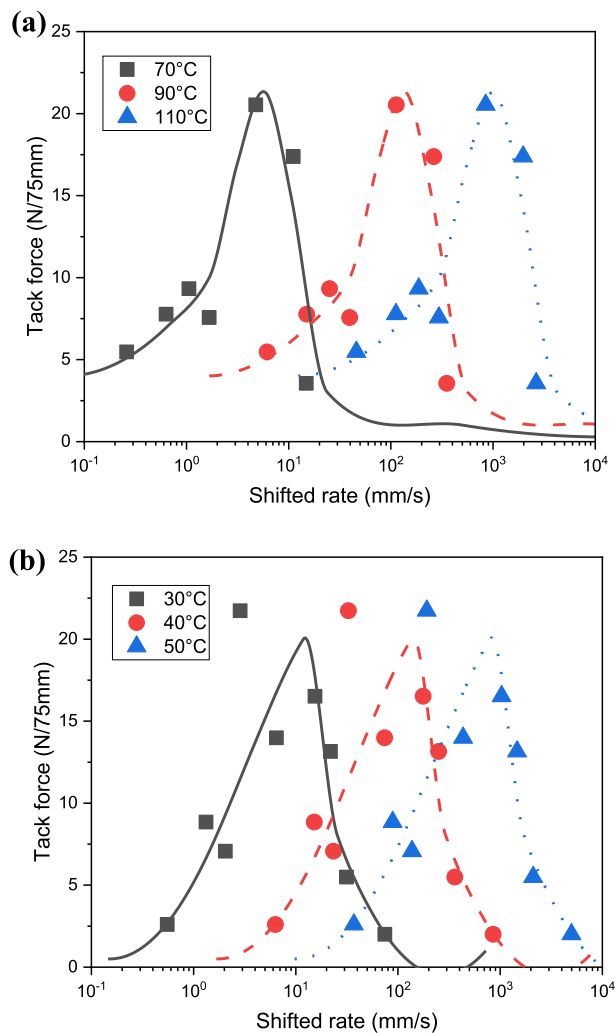
Fig. 5. Tack forces with respects to their shifted feed rates for (a) the solid resin system (at T_0 = 70 °C, 90 °C and 110 °C) and (b) liquid epoxy resin system (at T_0 = 30 °C, 40 °C and 50 °C). The curves connecting the measured tack values have been added to assist visualisation only (no best fit applied). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Flexural properties of 0°/90°/0° cross-ply carbon fibre composites manufactured with solid and liquid epoxy resins.

Matrix state at 25 °C	Accelerator content [wt. %]	Manufacturing process	Fibre volume fraction, V_f [%]	Flexural strength [MPa]	Flexural modulus [GPa]
Liquid	0	Bag press	55 ± 1	1116 ± 86	69 ± 5
Solid	0	Bag press	49 ± 1	1127 ± 183	74 ± 7
Solid	1	Bag press	46 ± 2	1070 ± 144	61 ± 9

4. Conclusions

A printing technique using a powder dispenser was developed to manufacture carbon fibre preforms using lower-cost solid epoxy resin formulations. These resins were formulated by mixing a solid epoxy resin and hardener at elevated temperature. Accelerator (1 wt%) was added to reduce the curing time of the resins from 8 h to 16 min. After
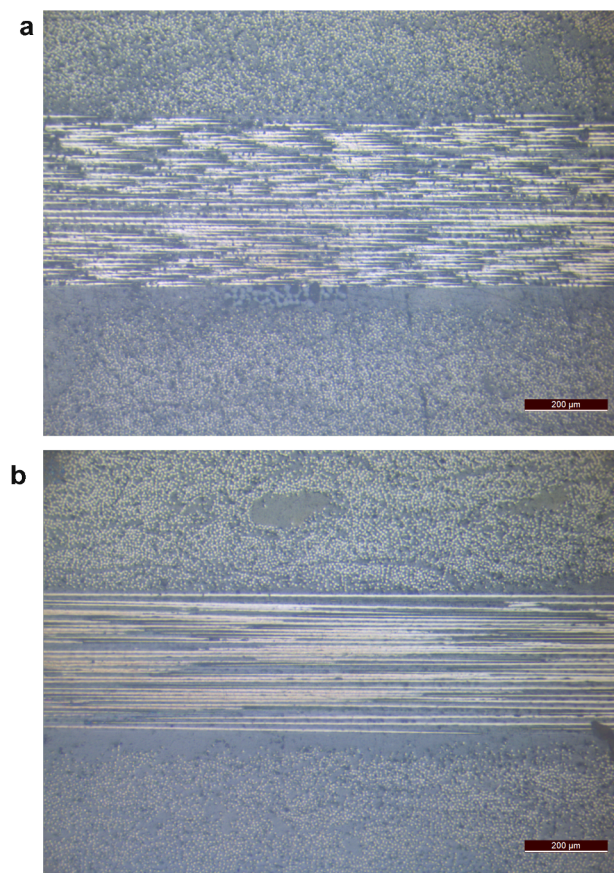

Fig. 6. Optical micrographs of microsections of carbon fibre reinforced composites cured without accelerator (a), and with 1 wt% accelerator (b). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

curing, the manufactured composites without accelerator had competitive flexural properties to composites produced using a conventional liquid epoxy resin. The flexural strength of composites manufactured without accelerator was 1127 ± 183 MPa, compared to 1070 ± 144 MPa for those with 1% accelerator. This could lead to the development of more affordable carbon fibre composites that could be used in a range of applications, such as marine and automobiles.

Furthermore, using tack testing we determined the temperatures at which it would be possible to deposit dry carbon tows to a resin film for both the solid epoxy resin system and a liquid resin benchmark. Despite needing higher temperatures for the solid resin compared to the liquid resin system, the solid resin was less sensitive to changes in temperature and feed rate. Optimum processing temperatures were determined for a low-cost ADFP method, where dry tows may be deposited onto a resin powder, which was dispersed beforehand using a printing head. These temperatures were determined by applying the time–temperature superposition (TTS) principle to rheology data and showed that temperature control during fibre placement is required to allow for increased deposition rates. The peak tack force can be shifted with respect to deposition rate, from 100 mm/s to 800 mm/s if the temperature is increased from 90 °C to 110 °C for the solid resin system. This temperature range is broader than for the liquid resin benchmark (40 °C to 50 °C), offering greater flexibility in terms of process temperature control.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.compositesa.2020.106205.

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