

# MANUFACTURING HIGH PERFORMANCE COMPOSITES USING SOLID EPOXY RESINS

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## Abstract

We aim to lower the cost of composite manufacturing by formulating solid epoxy resins. Cured solid epoxies are typically used as protective coatings e.g. in marine applications, and are in general much cheaper than the liquid epoxy resins used in composite production. The development of a cost effective composite manufacturing technique using cheaper starting materials, such as solid epoxy resins, will increase the likelihood of composite uptake in sectors where cost and feasibility have previously limited their adoption.

Powdered, uncured solid epoxy resins were formulated by mixing solid epoxies, hardeners and accelerators at elevated temperature. The resulting mixtures were then ground into powders that were applied directly onto carbon fibres. The fibres were then heated, infused with the melted resin system and subsequently cured. The following laminates have competitive mechanical properties when compared against other high performance composites. The laminates have a flexural strength and modulus of 919 MPa and 70 GPa, respectively. Using the same fibre format with a liquid epoxy resin results in a composite with a flexural strength and stiffness of 940 MPa and 80 GPa, respectively. Demonstrator 3D composite panels have been produced showing that is possible to mould the laminates during manufacturing.

## 1. Introduction

There is currently a considerable amount of worldwide effort to reduce the cost of composite component manufacture, from attempts to reduce the cost of the raw fibre through novel pre-cursor materials, to automation of the manufacturing process [1]. Much of this has been led by the aerospace industry, resulting in technologies such as Automated Tape Laying (ATL) and Automated Fibre Placement (AFP) [2]. However, these processes have mostly considered the application of thermoset or thermoplastic prepreg materials in the form of wide (ATL) or narrow (AFP) tapes. The former technique is suitable for large, gently contoured parts such as wing skins or fuselage sections. For more complex geometries, AFP is preferred. However, material lay-down rates reduce from a few hundred kilograms per hour for ATL, to around 3 kg/h for AFP. Much higher lay-down rates are generally required for composite mass production. The preferred composite manufacturing method in many cases is RTM [3]. The main limitation of using RTM is the high cost of the preforming stage. This uses dry fibre pre-forms for which the ATL process is unsuitable, and the AFP process is in its infancy. European funded projects such as AUTOW and ADVITAC have concentrated on using AFP with heavily processed dry fibre “tapes” which, whilst suitable for aerospace applications, are too

expensive (and slow) for industrial use [4, 5]. Other techniques such as braiding and filament winding are capable of high material deposition rates, but are limited by geometry.

Huntsman has developed a new composite manufacturing technique known as Dynamic Fluid Compression Moulding (DFCM) [6]. Fast cure liquid epoxy resin systems are placed by hand over a stitched or woven carbon fibre preform which is then compression moulded for 90 s to infuse and cure the composite part. The use of rapid curing resins creates fast production times. There is little material waste after the process, however the technique currently uses expensive liquid resin systems. Further concepts for reducing the cost of composite manufacturing are based on discontinuous fibre materials [7]. Whilst good in terms of damage tolerance (due to their quasi-isotropic nature) and speed of deposition, the resulting composite materials cannot hope to achieve the mechanical performance of continuous fibre materials, particularly with regards to strength. Structural components manufactured using these methods will inevitably be heavier than necessary.

Here we describe an affordable composite manufacturing method using solid epoxy resins. These resins are much cheaper than the liquid epoxy resins typically used in composite manufacturing.

## 2. Experimental details

### 2.1 Materials<sup>1</sup>

Solid epoxy resin and accelerator were supplied by Huntsman (Duxford, UK). Powdered hardener was supplied by AlzChem (Trostberg, Germany). Formax (Narborough, UK) provided the continuous carbon fibres.

### 2.2 Formulating solid epoxy resins

The epoxy and hardener were mixed together at room temperature by shaking them together in a sealed container. The epoxy and hardener were then mixed for 5 mins at 80°C using a Plastograph (Brabender, Duisburg, Germany) equipped with a mixer (W50EHT – 3 zones) at speeds ranging from 5-100 rpm. The same resin but containing accelerator was also formulated using this procedure, except the temperature during mixing was kept at 65°C to prevent premature curing. The solid resins were collected and stored below room temperature. The mixed resin was then ground into a powder using a pestle and mortar and sieved using a superfine mesh (0.2 mm aperture size) to obtain a relatively uniform particle size of approximately 25 µm.

### 2.3 Manufacturing 2D carbon fibre reinforced composites

Release agent (Frekote 700-NC, Henkel, Germany) was spread onto the surface of a hotplate (HP 1836-AH, Wenesco Inc., USA) in an area outlined with masking tape. The release agent was allowed to dry for 10 minutes and the masking tape removed. Vacuum bag sealant tape (Airdam 1, Airtech, Luxembourg) was placed outside the release agent coated area, where the masking tape had been. Four batches of resin powder, were prepared along with three layers of carbon fibre.

High temperature polyimide release film (Upilex-25S, UBE, Osaka, Japan), slightly larger in size than the carbon fibre layers, was placed onto the surface of the hotplate. On top of this release film, epoxy resin was distributed evenly through a superfine mesh (0.2 mm aperture size) over an area the size of a carbon fibre layer. The first fibre layer was then placed onto the resin. The next resin batch was spread through the sieve onto the carbon fibres, followed by another fibre layer, resin, the third fibre layer and the final resin batch. Polyimide release film, the same size as the one used previously, was placed on top of the last epoxy layer.

Vacuum bagging film (Easy Composites, UK) was pressed against the sealant tape to create a sealed vacuum bag. A through-bag connector with ¼turn valve and hosetail barb fitting (Easy Composites, UK) was placed at one end of the bag away from the sample and was used to connect the vacuum bag to a vacuum pump (Composites Vacuum Pump, Easy Composites, UK).

The vacuum pump was switched on to evacuate the bag. A metal plate was placed above the sample and the specimen slowly heated to 150°C using a heating plate. The vacuum valves on the pump and the through-bag connector were then closed to maintain the vacuum and the vacuum pump switched

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<sup>1</sup> This technology is in the process of being patented and therefore certain details have to be omitted.

off. Composites without accelerator were cured for 8 h at 150°C. Samples containing accelerator were cured at 150°C for 30 mins (the vacuum pump was switched on for the entire curing process to encourage the removal of entrapped air).

#### 2.4 Manufacturing 3D carbon fibre reinforced composites

A different manufacturing procedure to that described in section 2.3 (2D composite production) was used. Resin powder was melted onto three carbon fibre layers at 65°C and then cooled. An aluminium block (65 mm, 37 mm, 6 mm) was wrapped in blue release film (A6000 blue 12 micron P3 perforated release film, Cytec, Wrexham, UK) and placed above a layer of polyimide release film. The fibres containing resin were then placed over the aluminium block. Finally, a 1mm thick silicone release film (FenForm, Cured S202 Silicone, Fenner Precision, Lincoln, UK) was draped over the laminates and a vacuum bag was used to enclose the setup. Curing was then performed as described in section 2.3.

#### 2.5 Preparation of DMTA and flexural test specimens

Cured composite samples were cut into specimens for DMTA (50 mm × 10 mm) and flexural testing (80 mm × 10 mm) using a water cooled diamond bladed cutter (Diadisc 4200, Mutronic, Rieden, Germany). For each sample the 0° direction was parallel to the longer specimen dimension.

#### 2.6 DMTA of composites

The viscoelastic properties of the composites were analysed using DMTA on an RSA-G2 (TA Instruments, Eschborn, Germany). The tests were performed in 3-point bending at frequency of 1 Hz. The samples were heated at 5°C/min from 30°C to 140°C.

#### 2.7 Flexural testing of composites

The composite samples were analysed using three point bending tests on an Instron 5969 (Bucks, UK) equipped with a 50 kN load cell. The tests, in accordance with ASTM D7264-07, were performed at a crosshead speed of 1 mm/min and 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:

$$E_f = (L^3 m)/(4bh^3). \quad (1)$$

where  $m$  is the gradient of the linear section of the load - displacement curve,  $L$  is the support span,  $b$  the beam width and  $h$  the beam thickness. The flexural strength of the composites was calculated using Eq.2:

$$\sigma = 3PL/(2bh^2). \quad (2)$$

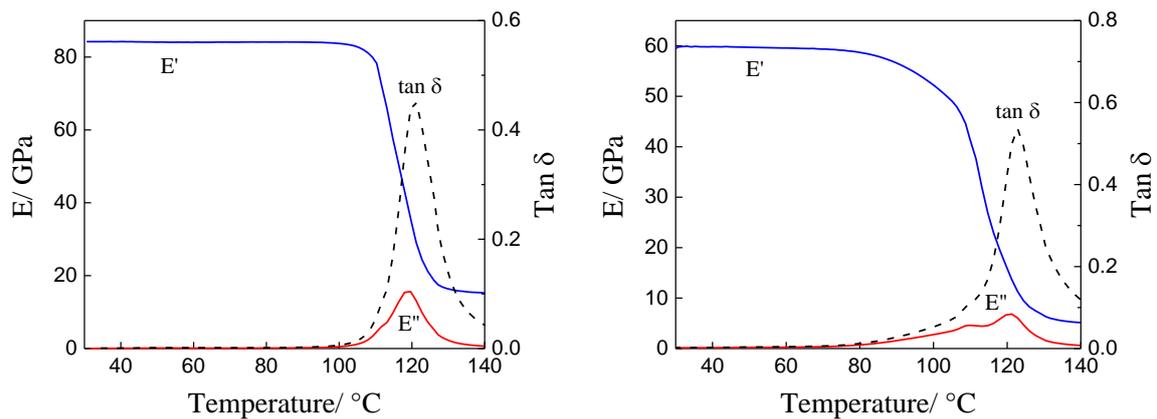
where  $\sigma$  is the maximum stress at the outer surface at the mid-span of the specimen (MPa) and  $P$  the applied force at failure (N). The average of at least five specimens was taken to calculate the flexural modulus and flexural strength of each sample.

### 3. Results and Discussion

#### 3.1 Viscoelastic properties of composites

The composite manufactured without accelerator had a high storage modulus ( $E'$ ) of 84 GPa at 30°C (Fig. 1, left). The  $T_g$  of the resin was 120°C (peak in  $E''$ ) and at this temperature the storage modulus of the composite had dropped significantly due to softening of the resin.

The composite manufactured with accelerator had a storage modulus of 60 GPa at 30°C and a  $T_g$  at 120°C (Fig. 1, right). The lower storage modulus (stiffness) of this composite at 30°C, compared to the composite without accelerator, can be explained by its higher void content. Voids were present in all the composites manufactured and this was most likely a result of the resin gelling before it had time to fully degas. However, the composite manufactured with accelerator had a significantly higher void content than the composite without accelerator. This can be explained as the resin containing accelerator gelled faster and at a lower temperature, therefore limiting time for air removal during preform infusion.



**Figure 1.** DMTA of a composite cured without accelerator (left), and with accelerator (right).

#### 3.2 Flexural properties of composites

The flexural properties of the composites are summarised in Table 1. The flexural strength and modulus of the composite manufactured without accelerator were  $919 \pm 62$  MPa and  $70 \pm 3$  GPa, respectively. In comparison, the composite manufactured with accelerator has a slightly lower flexural modulus ( $64 \pm 6$  GPa) and a significantly lower flexural strength ( $675 \pm 143$  MPa). This was due to the high void content in the composite.

**Table 1.** Flexural properties of composites manufactured with solid and liquid epoxy resins.

Matrix state at 25°C	Accelerator content	Manufacturing process	Flexural strength (MPa)	Flexural modulus (GPa)
Liquid	No	Infusion	940	80
Solid	No	Melt infusion	$919 \pm 62$	$70 \pm 3$
Solid	Yes	Melt infusion	$675 \pm 143$	$64 \pm 6$

The flexural properties of a composite manufactured using the same fibre format but with a liquid epoxy resin system is compared. The composite has a flexural strength and modulus of 940 MPa and 80 GPa, respectively [8]. These values are similar to the composite manufactured without accelerator and indicate that we are able to produce high performance using our new manufacturing method.

### 3.3 3D composite production analysis

An image of the manufactured 3D composite is shown in Figure 2. The composite forms and cures well around the metal mould. From our trial-and-error experiments a 1mm thick silicone film creates the best finish, although further experimentation is required to confirm this.



**Figure 2.** 3D composite cured with accelerator.

## 4. Conclusions

A novel, low cost carbon fibre composite manufacturing method, using solid epoxy resins, has been developed. Solid epoxy resins with cured thermal properties suitable for high performance applications were formulated. An accelerator was added to reduce the curing time of the resins from 8 hours, without accelerator, to less than 10 mins with accelerator. The solid, powdered resins were infused into carbon fibres using melt infusion and then cured. The resulting composites have competitive flexural moduli when compared with composites manufactured from liquid epoxy resins. However, the resins containing accelerator suffer from high void content as they gel rapidly during the infusion process thus limiting air removal. The void content significantly lowers the flexural strength of the composites. For example, the composite manufactured with accelerator (high void content) had a flexural strength of  $675 \pm 143$  MPa, whereas the composite manufactured without accelerator (low void content) had a higher flexural strength of  $919 \pm 62$  MPa. Despite the presence of voids, there are still many applications for these affordable, lightweight composites.

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