FIBRE REINFORCED POLYIMIDE COMPOSITES AND STRUCTURES MANUFACTURED WITH RESIN TRANSFER MOULDING – OVERVIEW OF PROCEDURES AND PROPERTIES

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Abstract

In this paper, the major outcomes from a recently completed research program with ambition to develop polyimide carbon fibre composites with temperature ability above 360°C are reported. Data from characterisation of the processing properties such as viscosity and cure behaviour are presented alongside with data on the mechanical properties at room temperature of quasi-isotropic composites based on the developed resin and 8-harness satin weave carbon fibre fabrics. The paper also contains a demonstration of the use the material system in a demonstrator component.

1. Introduction

Although ceramic and metal matrix composites can be used at several thousand degrees, there are strong incentives to increase the use of fibre reinforced polymer composite (FRPC) in high temperature applications primarily since they are more lightweight, have better fatigue properties and are more ductile [1]. A major limitation of traditional FRPC e.g. based on epoxies however is the poor ability of the matrix to maintain load transfer capability at high temperatures as well as their relatively poor fire resistance.

In response to this the current communication present in summary the results from a development program *HicTac - High performance composites for demanding high Temperature applications*, with focus to develop thermosetting polyimide formulations and the associated manufacturing methods [2]. Throughout the work we are using a phenyl ethynyl terminated polyimide formulation NEXIMID® MHT-R (Nexam Chemicals AB, Sweden). The resin is specially developed in the program by Swerea SICOMP AB and Nexam and combines extreme temperature stability with attractive processing characteristics, enabling cost efficient manufacturing using resin transfer moulding (RTM). The current paper will summarize the major outcomes from previous reported work as well as to present and demonstrate how the material system is used to prepare real demonstrator components with geometrical similarities to a fan blade structure.

2. Materials

The resin used in the work was a thermosetting polyimide NEXIMID® MHT-R, exclusively formulated for the HicTac-project by Nexam Chemical AB, Sweden (noted MHT-R hereafter) [3]. This thermosetting polyimide is prepared by a combination of 6F-dianhydride (6-FDA) backbone, 4-(Phenylethynyl)Phthalic Anhydride (4-PEPA) end-group crosslinker and ethynyl bis-phthalic

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anhydride (EBPA) main chain crosslinker. The resin was engineered to fulfil two major requirements: the glass transition temperature, T_g, of the cured system should exceed 360°C and the resin should exhibit processing characteristics that permit RTM manufacturing of composites. The compatibility of the resin with RTM is illustrated in Figure 1 (from [4]). It can be seen that the once the resin is molten (occurs at around 200°C) the melt viscosity reduces with increasing temperatures to values between 200 mPas and 400 mPas at temperatures between 280°C and 320°C. The increase in viscosities above these temperatures indicates that a polymerization reaction is initiated above these temperatures. The observation of curing at temperatures above 320°C is further supported by the dynamic differential scanning calorimetry scans (DSC) presented in in Figure 2. Thornel® T650/35 8-harness satin woven fabric (370 g/m^3) supplied by Sigmatex (the fibres were produced by CYTEC were used to prepare the composites and demonstrators.

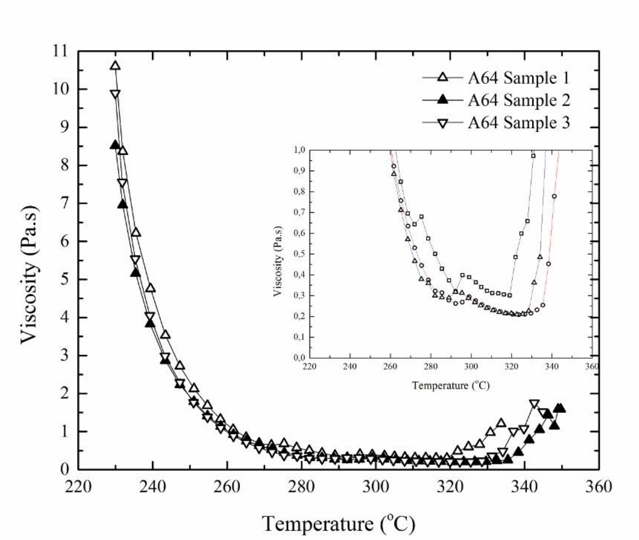

Figure 1 Shear viscosity vs. temperature for MHT-R (resin is denoted Nexamite A64 in original source [4])

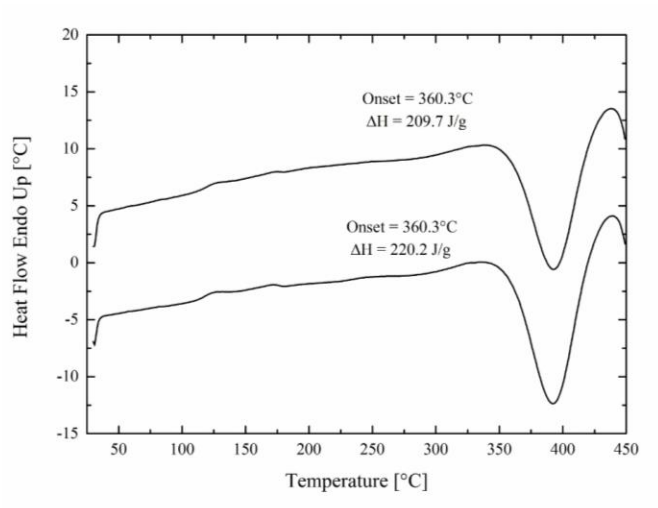

Figure 2 DSC-scan from 30 to 450°C at 10°C/min for MHT-R/Nexamite A64 [4].

3. Processing and properties

RTM was the preferred composites manufacturing method in the project. 8-harness satin weaves were, used to prepare composites with quasi-isotropic $[(-45/45)/(0/90)]_{2s}$ lay-ups. Flat laminates prepared in a 350 mm × 350 mm electrically heated steel tool were used to assess manufacturing schemes and to prepare specimens for subsequent mechanical testing. A flow and pressure controlled injection piston

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supplied by ISOJET was used for injecting the resin during RTM-manufacturing. The curing of the laminates was typically carried out in the following steps: melting and homogenization of the polyimide in the piston at 240°C for 30 minutes; degassing using vacuum at 240°C for 10 minutes; resin injection at 290°C; curing at 320°C and 12 Bar for 2 h; temperature increase to 370°C at a heating rate of 1.7°C/min, curing at 370°C and 12 Bar for 2 h, cooling down to 80°C during 12 h and demoulding [5]. A cure according to the mentioned scheme yielded in composites with typical Tg of around 370C. The influence of post-cure was evaluated in a separate study [6]. The results from this study demonstrated that T_g could be increased towards 400-440°C with a free standing post-cure, see Figure 3. Besides increased T_g, the post-cure also caused limited weight loss due to thermal degradation and an increase in internal damage in the form of transverse microcracking.

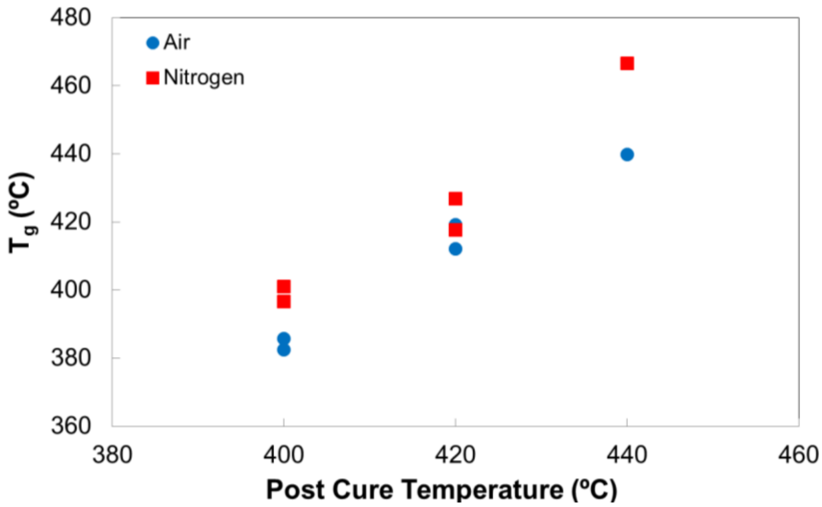

Figure 3 Influence from temperature and environment on Tg after 2 hour post-curing [6].

Figure 4 Representative cross-section from the laminate acquired by optical microscopy [5].

Mechanical testing was performed on samples prepared from the 350×350 mm multidirectional laminate mentioned in previous section. A micrograph showing the microstructure of a typical cross section of a test sample is presented in Figure 4. Results from mechanical tests at room temperature were presented in previous reports [3] and are summarized in Table 1. The tensile testing results were obtained from tests on three specimens and according to ASTM D3039 at a constant cross-head speed of 1 mm/min using an Instron 5801 servo-hydraulic machine equipped with a 100 kN load cell. The compression testing was performed using the classic Wyoming modified ITRII fixture. Five samples were tested. For the short-beam shear test, eight specimens with dimensions 18 mm ×6 mm were tested in three-point bending with a 4:1 span-to-depth ratio.

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Table 1. Mechanical properties from tests at room temperature (from [5])

Property	T650/MHT-R
Tensile Stiffness (GPa)	48.5±0.6
Tensile Strength (MPa)	471.0±36.8
Tensile Strain-to-failure (%)	1.05±0.10
Compressive Stiffness (GPa)	44.8±2.7
Compressive Strength (MPa)	371.3±14.8
Compression Strain-to-failure (%)	0.86±0.08
ILSS (MPa)	47.7±6.7

4. Demonstration of feasibility for upscaling

The potential of using the developed materials in components with more complex geometries was also assessed in the project. A generic geometry with approximate dimensions 300 mm x 150 mm x 8 to 30 mm (see Figure 5) was used for the purpose. The manufacturing of the demonstrator components were conducted in heated metal tools according to the same procedures described in previous section. In summary the results were very encouraging, the demonstrators were of high quality.

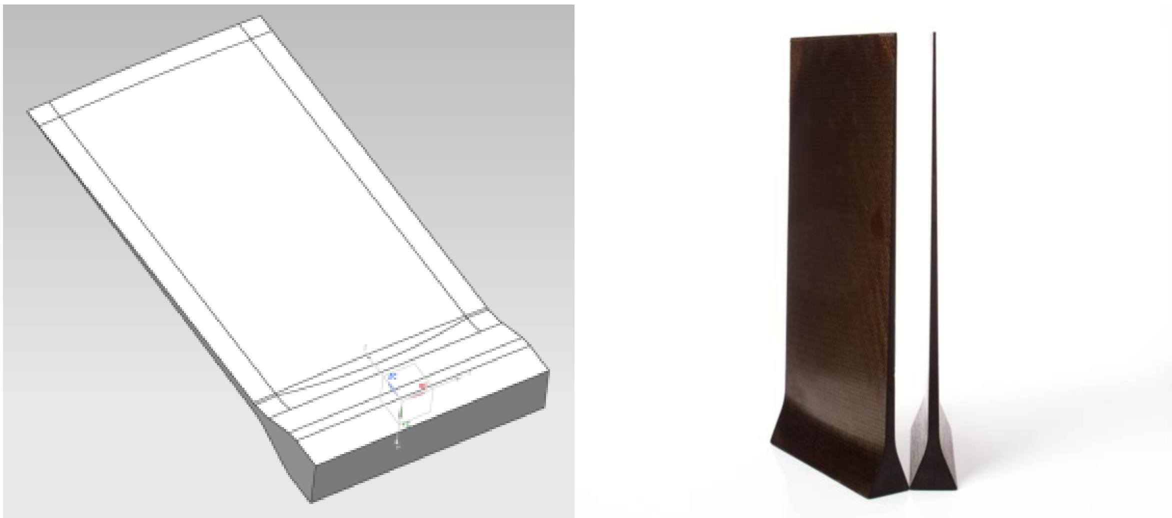

Figure 5 Illustration showing CAD-drawing of the demonstrator and photograph of two manufactured high temperature carbon fibre demonstrator components.

4. Summary

The goal to develop a new polyimide formulation and establish that the system can be used with RTM-manufacturing was successfully achieved within the project. It has been shown that the resin has sufficiently low viscosity to enable RTM-manufacturing and that the resulting composites has temperature ability, in terms of resin T_g, that exceeds the originally anticipated 360°C. Mechanical tests on quasi-isotropic laminates yields in results that are acceptable and comparable with data in literature for similar systems [7-9]. Finally it was established, by manufacturing demonstrator components, that the material system can readily be used to manufacture real components.

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References

[1] S. Black Are high-temp thermosets ready to go commercial? *High-Performance Composites, November, 2004*.
[2] HicTac – High performance composites for demanding high Temperature applications, CleanSky – *Research and Technology Development Projects, Grant agreement SP1-JTI-CS-2012-02*, 2012.
[3] Nexam Chemical, NEXIMID® MHT-R, Product Data Sheet, 2016.
[4] Fernberg P, Gong G, Mannberg P, Processing and Properties of New Polyimide Composites with High Temperature Ability, 16th European Conference on Composite Materials, ECCM16, Seville, Spain, 2014.
[5] Tsampas SA, Fernberg P, Joffe R, Mechanical Performance of Novel High Tg Polyimide Matrix Carbon Fibre Reinforced Laminates, 20th International Conference on Composite Materials, ICCM20, Copenhagen, Denmark, 2015.
[6] Fernberg P, Joffe R, Tsampas SA, Mannberg P, Influence of Postcure on Carbon Fibre Polyimide Composites with Glass Transition Temperatures Above 400°C, 20th International Conference on Composite Materials, ICCM20, Copenhagen, Denmark, 2015.
[7] JW. Connell, JG. Smith Jr., PM. Hergenrother, JM. Criss, High temperature transfer molding resins: Composite properties of PETI-330, *International SAMPE Technical Conference Series. vol. 48 Covina, CA, 2003*, pp. 1076-1086.
[8] Kakuta Y. *et. al.*, Axial Fatigue Strength of Open-Hole Specimens of T800/PMR-15 Carbon-polyimide Composite at Room and High Temperatures, *In Proceedings of International Conference on Composite Materials (ICCM-18)*, South Korea, 2011.
[9] S. Bain, H. Ozawa, JM. Criss Jr., Mechanical properties of thermally aged peti-330 laminates fabricated by resin transfer molding. In Proceedings of International SAMPE Symposium and Exhibition, (2007), v 52, 15 p.

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