IMPROVEMENT OF THE TOUGHNESS OF EPOXY RESIN SYSTEMS USING THERMOPLASIC BINDERS

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ABSTRACT

This paper describes the changes in the impact fracture toughness of carbon-epoxy resin composites produced by means of thermoplastics distributed in powder form between the individual fibre layers. The influences of the type and amount of polymer and its grain size, as well as of combinations of different thermoplastics, are considered and discussed. Variations in the physical form of the powder make it more difficult to predict its distribution after resin injection and for this reason the hand layup process was used to produce the composite components. Thus during the curing cycle in the autoclave the thermoplastics may display dissolving, melting or non-melting behaviour. The resulting samples were tested and evaluated according to the compression after impact standard, and this showed that the melting type of polymer led to the greatest improvement, and that the impact fracture toughness could be significantly increased with a polymer addition of only 6 g/m^2 of component surface area.

MATERIALS

The reinforcing layers of the epoxy laminate were built up from a unidirectional carbon-fibre fabric of specific weight 298 g/m^2. This fabric was based on Toho Tenax® J IMS60 E13 fibres supplied by SAERTEX GmbH. Two single-component resin systems, HEXFLOW RTM 6 from Hexcel, and Cycom 890 RTM from Cytec, were used, both applied in the as-delivered condition. The polymers used for modifying the impact toughness were made available by EMS-Griltech in powder form. These were: Phenoxy 85 and Phenoxy 95, Phenoxys with medium and high molecular weight respectively; CoPA 130, CoPA 180 and CoPA 300, Copolyamides with different glass transition temperatures respectively.

LAMINATE MANUFACTURING

A central goal in preparing these laminates was to ensure a uniform distribution of the polymer particles in the interlaminar layer. Because it was not possible to draw unambiguous conclusions as regards the effect of the resin infusion process itself on this distribution, the samples were prepared by hand lay-up. For this purpose steel plates treated with the release agent Frekote 770-NC were used for the tooling (see Figure 1). Both tool and resin were preheated to 80°C. The quasi-isotropic fabric lay-up was 16-ply $[0/+45/-45/90]_{2S}$, this material being cut into 330mm squares.

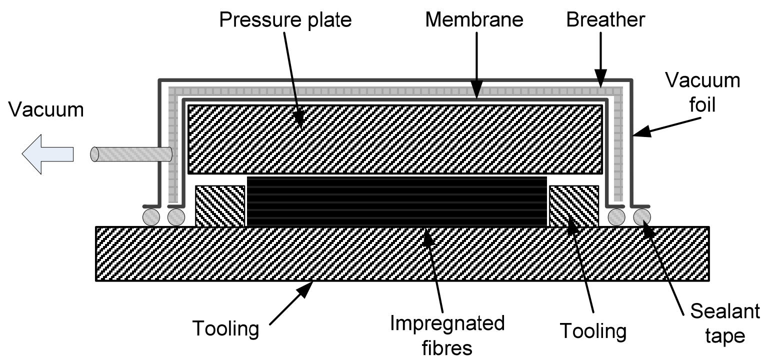

Figure 1: The hand layup assembly used for producing the fibre-reinforced composite test plates

A finely meshed sieve was used to sprinkle a uniform layer of thermoplastic powder at defined surface loadings of 6, 12 or 18 g/m^2. The fibre packet was closed off with a pressure plate which served to ensure a flat upper surface, a semi-permeable membrane preventing the resin from escaping from the impregnated fibre packet. The resulting assembly was placed in the autoclave, which was then evacuated and heated up to 120°C at 1.5°C/minute and held at that temperature for 30 minutes, after which the pressure inside the autoclave was raised to 6 bar. The temperature was then raised to 180°C at the same rate of 1.5°C/minute, held at this level for 90 minutes in the case of RTM6 and 120 minutes for Cycom 890, and then slowly reduced to ambient temperature and pressure. All the resulting composite laminated samples were tested according to the AITM 1-0010 (ref. 1) standard, for homogeneity, delamination and porosity. This was done by means of ultrasonic phased-array scanning using the Olympus OmniScan MX.

EXPERIMENTAL

Compression after impact (CAI) samples were cut out of the composite laminates with a Mutronic DIADISC 5200 water-cooled diamond saw, to an average thickness of 4.1mm, 100mm wide by 150mm long. The subsequent impact loading of these samples, the measurement of the depth of the dent and the analysis of the damaged surface, as well as the compression test, were all in conformance with the AITM 1-0010 standard. The procedure was to clamp the sample to a heavy solid plate and to subject it to the impact of a 4.45 kg drop weight with a hemispherical nose 16mm in diameter, an anti-rebound device ensuring that no second impact took place. The impact energy was 30 J for all the samples of the same test. After the depth of the dent was ascertained the samples were again subjected to ultrasonic testing, in which the delamination zone was defined using the half-value method and then quantified numerically and recorded. The compression testing was performed using a Schenk-Trebel RM 250 materials testing machine with a constant test speed of 0.5 mm/min and standardised tooling. Six samples were produced and tested for each type of composite laminate.

RESULTS

The volume content of all the composites tested was between 60% and 73%, with an average value of 67% for all the samples tested. In order to facilitate comparison between samples, all the compressive forces mentioned in this study have been normalised to that average of 67%. Literature reference (ref. 2) provides a detailed description of the normalising procedure used.

In all, two different series of tests were carried out, both of them using the RTM6 and Cycom 890 resin systems, to investigate the influence of various material parameters on the impact damage results, these parameters including the type of thermoplastic, as well as the specific amount of the thermoplastic used. First of all samples were produced without any thermoplastic addition, to serve as a reference defining the lower limit, that is consequent on the worst damage from impact loading, as well as the upper limit (undamaged samples) of the residual compressive strength. In Test Series 1, two grams of the relevant thermoplastic powder was distributed between the individual fibre layers, corresponding to a specific loading of 18 g/m^2 (see Figure 2, in which the height of the column shows the average and the bar the spread over the six samples; this is true also of Figures 3 and 4). It can be seen that the addition of all thermoplastic powders brought an increase in residual compressive strength, with CoPA 130 and CoPA 180 showing the best improvement in performance. As with residual compressive strength, the polymers CoPA 130 and CoPA 180 produced the best results, followed by the Phenoxy, which shows no great difference, and the CoPA 300, which falls somewhere between the Phenoxy and the unmodified samples. As regards the differences between the two resin systems, they both behave in the same way, although Cycom 890 systematically shows the better results. This is particularly striking for samples modified with 18 g/m^2 of CoPA 180, which attain more than 100% increase in the residual compressive strength.

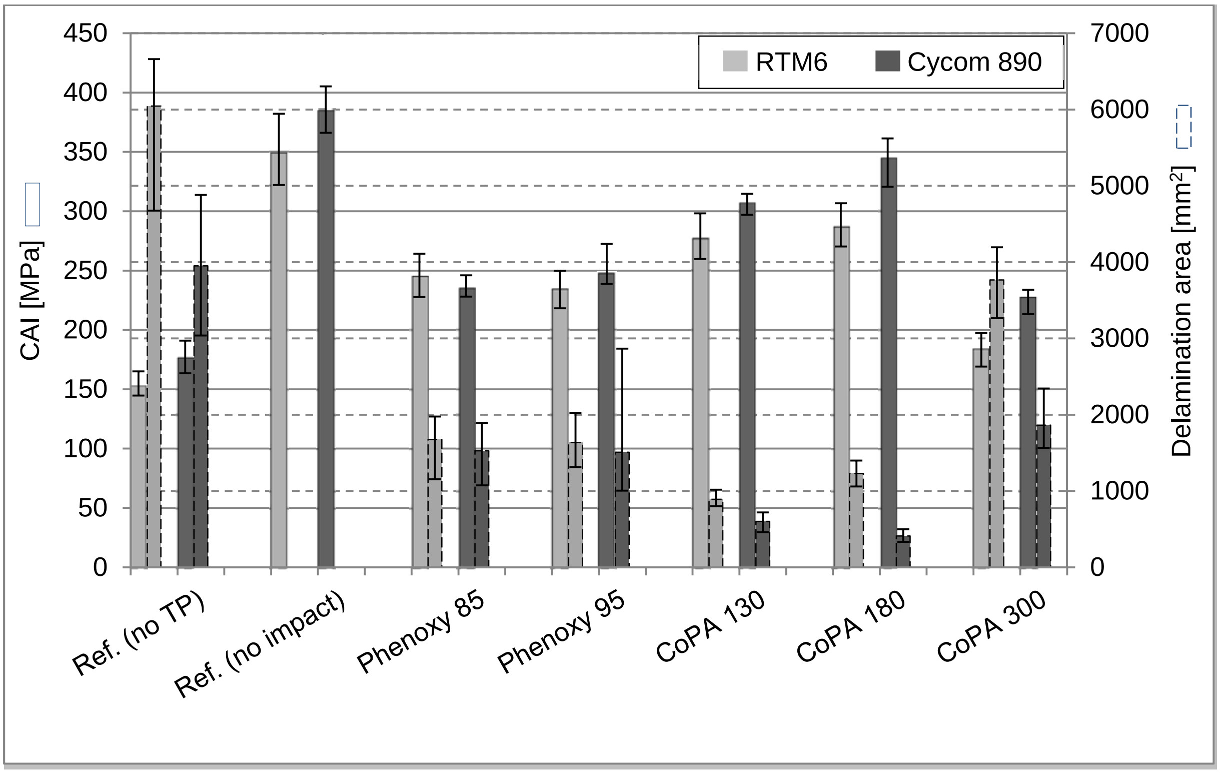

Figure 2: The effect on residual compressive strength and delamination of the addition of 18 g/m^2 of thermoplastic in the interlaminar layers

Based on the results of Series 1 the number of polymers investigated further was limited to those showing the best potential so far. These were CoPA 130 and CoPA 180. Series 2 considered the effect of different specific thermoplastic powder loadings. Accordingly, first 0.65 g and then 1.3 g of powder were distributed between the individual fibre layers, corresponding to specific loadings of 6 and 12 g/m^2. The resin systems were RTM6 and Cycom 890 as before. Figures 3 and 4 clearly show that even with only 6 g/m^2 the delaminated area is reduced by more than 60%, while the residual compressive strength is increased by 50%. In Figure 3 it can be seen that for all combinations of resin system and thermoplastic modifier the residual compressive strength increases with increasing thermoplastic loading, but tends towards the maximum of the undamaged samples. The same pattern also occurs with the reduction in delamination (Figure 4).

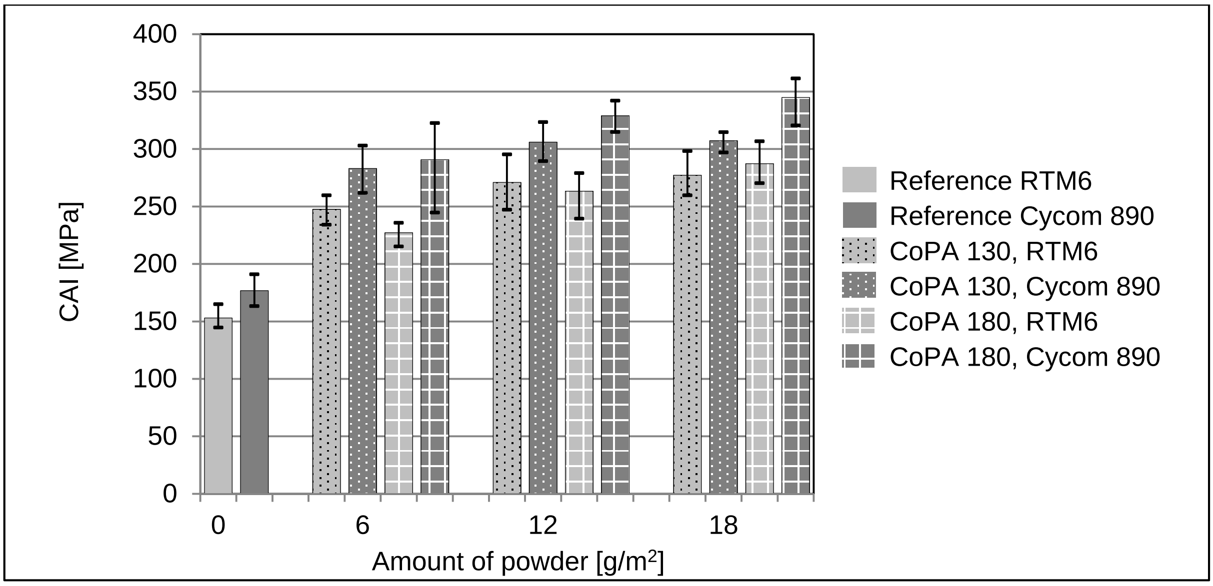

Figure 3: The effect on residual compressive strength of increasing the specific loading of thermoplastic powder.

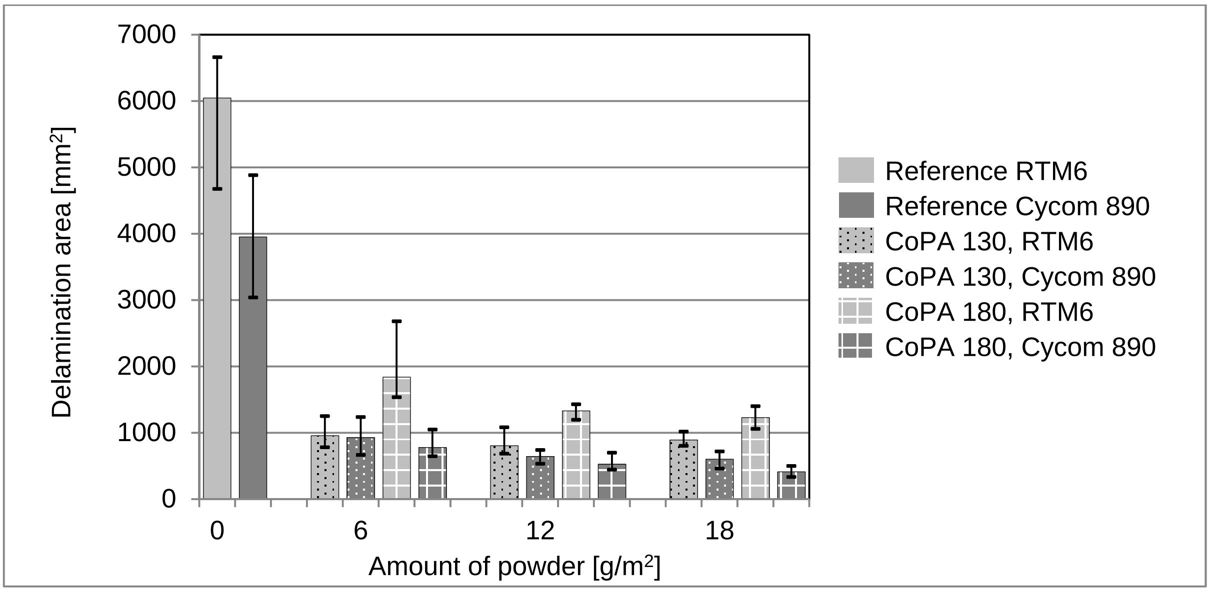

Figure 4: The effect on delamination of increasing the specific loading of thermoplastic powder.

ACKNOWLEDGEMENTS

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(ref. 2): Anon. Polymer matrix composites – vol. 1. Guidelines for characterization of structural materials. MIL-HDBK-17-1F, U.S. Department of Defense, 2002, Chapt. 6, p. 43.