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THE INFLUENCE OF HEAT TREATMENT AND FINISHING ON THE MECHANICAL PROPERTIES OF LAMINAR COMPOSITES

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ABSTRACT

The aim of this investigation was to define the optimum conditions of obtaining glass fabric - epoxy resin laminar composites with appropriate mechanical properties that satisfy the quality needed for production of printed circuit boards for micro-electronics. Commercial materials: domestic glass woven fabric, different types of silane finish and epoxy resin were starting materials in obtaining composites.

The conditions needed for the thermal removal of the original size (protective coatings) from glass fabric were investigated. The optimal heat treatment should be performed on temperatures less than 550°C, while cooling rate should be as slow as possible. In this manner the fabric has less than 0.1% of residual size, and the mechanical properties remain satisfactory.

Two most commonly used silane based finishes were applied on heat-treated glass fabric. Tensile strength of the composite material made of thermally and chemically treated glass fabric and epoxy resin was investigated. The possibility of using domestic glass fabric in production of printed circuit boards for microelectronics has been confirmed. Finish with amino functional group and lower heat treatment temperature should be used for obtaining glass-fabric epoxy resin laminar composites with desirable mechanical properties.

Key words: Heat treatment, silane finishes, tensile strength, glass fabric - epoxy resin laminar composites

INTRODUCTION

Due to the combination of superior mechanical and dielectric properties, epoxy resin - glass fiber composites are becoming increasingly important for many modern technologies, including electronics and high and medium voltage electric plants [1, 2].

The role of the finishing is to enable complete wetting of the fiber by the matrix resin, which results in the efficient stress, strain and voltage distribution from the matrix to the fibers [3-6]. Newly drawn fibers are normally coated during drawing with a "size", a thin layer of substance that protects the fiber from damage and undesirable environmental interactions. This size is ordinarily removed prior to composite fabrication and replaced with a "coupling agent" or finish that promotes better bond between fiber and matrix [7]. The original coatings in woven fabrics are so-called "textile emulsions" which consist of a lubricant that makes the fibers smooth and protects them from internal friction during manipulation (hydrogenated vegetable and mineral oils), an adhesive for connecting the filament (dextrin, gelatin, poly(vinyl alcohol), poly(vinyl acetate), poly(butyl acrylate) etc.) and an emulsifier that stabilizes the emulsion (non-ionogenic and anion active emulgators). This type of coating enables the sliding of fiber, disables the dispersal of multifilament fibers in to separate filaments and protects against moisture absorption.

It is necessary to remove the textile emulsion before glass fabric becomes the part of the composite structure. The removal can be performed by chemical means, but it is more common to perform appropriate heat treatment. The residual after such treatment is approx. 0.2% of the lubricant, but it is also possible to obtain less than 0.1% [8].

The other type of coating applied during surface treatment of glass fibers are adhesive promoters or finishes applied in fiber reinforced composites. These coatings have important role to form a bond between organic polymers of matrix and reinforcing inorganic materials such as glass fiber or glass fabric. Therefore surface treatment is performed with materials that posses the property of reacting with inorganic glass as well as organic polymer, forming strong bonds between them. The modified silanes with different functional groups are widely used today as finishes [9,10].

The optimum conditions of heat treatment of glass fabrics as well as selection of appropriate finishes for obtaining glass fiber - epoxy resin laminar composites with mechanical properties suitable for printed circuit boards in microelectronics were investigated. First the optimal heat treatment condition for removal of coating from woven glass fabric were established. Than two types of finishes based on silanes were applied on heat-treated fabrics. The quality of the composite material made of thermally and chemically treated glass fabric and epoxy resin was controlled by measuring the tensile strength of composite.

EXPERIMENTAL

The commercial glass woven fabric (Tehnotex, Sombor, No. 558) made of E glass fibers was used for the investigation. The fabric had specific mass of 200 g/m^2; set of warp was 164 h/dm, while the set of weft was 140 h/dm.

Mechanical properties of the heat-treated fabrics were investigated in order to obtain the precise criteria for the temperature regime used for coating removal. The tensile strength of heat treated fabric was examined both for warp and weft. For each set of experiments, ten 100 mm x 50 mm samples were used. The mean value of experimental results is given. The measurements were performed on textile fabric tester TextTest Inc., Switzerland under 500 kg load. The instrument directly shows the braking force and relative elongation.

Different heating regimes were investigated in order to define the most appropriate to remove the original coating from woven glass fabric. A Heraeus oven with a controlled heating rate was used for heating, while the temperature inside the oven was monitored using a HP 3497 Data Acquisition System.

Two types of finishes - Dynasylan® commercial organo-functional silanes were used on heat-treated glass fabrics. There were: 3-aminopropyl-triethoxy-sylane (with trade name AMEO) and 3-glycidyloxypropyl-trimethoxysilane (with trade name GLYMO).

Both silanes were previously hydrolyzed to form silanol that reacts with glass fabric, while the hydrophobic residue bonds to the epoxy matrix. The hydrolysates were prepared as a 0.4% solution. The hydrolysis time was 120 min at 20°C. The heat-treated glass fabrics are then immersed in silane solution, rotated for 5 min, finally freed from liquid by squeezing and drying at 150 °C for 20 min.

After thermal and chemical treatment the composites themselves were prepared. The epoxy resin Cy 233 (Ciba-Geigy) based on bisphenol A was used, together with HT 972 hardener (aromatic amine) and XB 3022 accelerator (tertiary amine). The layers in the laminar composite were laid manually. The composite consists of two layers of glass fabrics with silane coating embedded between epoxy resin layers. The glass fabric layers are positioned at 90° degrees one to another, which is the usual way of producing these laminates when used for printed circuit boards. The laminate thickness was 1 mm, while the glass fabric content was 60 % mass.

The samples obtained in described manner were subjected to the tensile strength investigation using an Instron instrument according to ASTM D 2290/64T [11].

RESULTS AND DISCUSSION

According to the TG diagrams it is clear that the coating is completely removed by 600 °C [12]. The following part of this study consisted of investigation of the mechanical properties of thermally treated fabrics that were not subject to surface processing. The influence of the cooling rate on the mechanical properties of heat-treated glass fabric was investigated. All the samples were heated at 5 °C/min, kept at 500 °C for 60 minutes and then cooled to ambient temperature at various cooling rates. Investigated heat treatment regimes are given in Figure 1.

Figure 2. shows the influence of the cooling rate on the breaking force of investigated fabrics, while the influence of the cooling rate on the relative elongation of fabric is given in Figure 3.

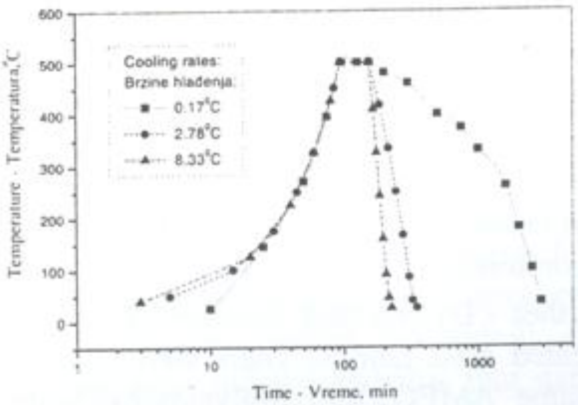

Figure 1 - Heat treatment regimes

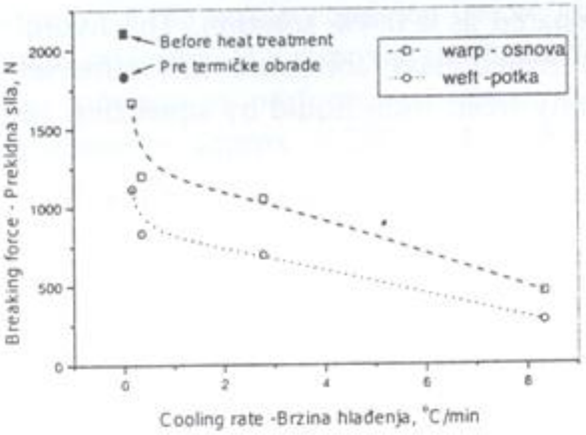

Figure 2 - Breaking force of the fabric vs. cooling rate

Slower cooling rates improve the mechanical characteristics tensile strength and relative elongation of the heat-treated samples. Therefore the further heat treatment was carried out with as slow cooling rate as it was possible to obtain.

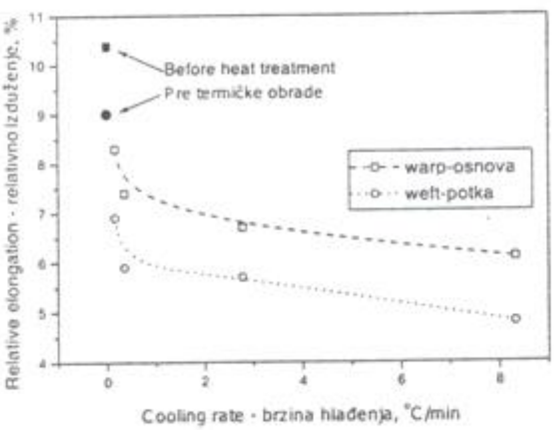

Figure 3 - Relative elongation vs. cooling rate

For next series of investigations fairly similar regimes of heat treatment were chosen, differing solely in maximal heating temperature (500 °C, 525 °C and 550 °C). The samples were heated at a heating rate of 5°C/min, kept at the maximum temperature for 60 minutes and slowly cooled (0.17 °C/min) to ambient temperature.

The obtained samples were exposed to surface treatment with two selected silane coatings giving six type of samples, which were used for making epoxy matrix – glass fabric composites. The results of the investigation of the mechanical properties of these composites are shown in Figure 4. The type of composite is named after the trade name of finish used for its preparation.

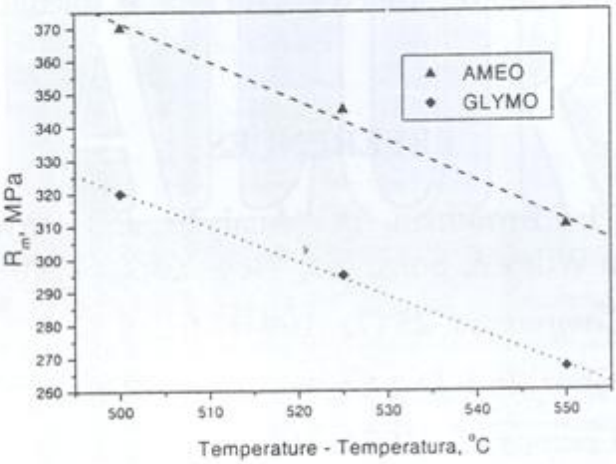

Figure 4 - Tensile strength (Rm) vs. heat treatment temperature and types of finishes

The tensile strength of composites linearly decreases with the increase of the heat treatment temperature. Composite samples with AMEO finish have, for all investigated heat-treatment regimes, from 13.5 to 14.5 % higher tensile strength than composites prepared with GLYMO finish. It was established that best mechanical properties of obtained glass-fabric epoxy resin laminar composites were achieved when heat treatment was performed at lowest investigated temperature, 500 °C, using AMEO finish.

CONCLUSION

By investigating of the mechanical properties of heat-treated glass fabrics, that did not perform surface treatment, the optimal regime of heat treatment was established. With that treatment the fabric has less than 0.1% of residual size, and the mechanical properties remain satisfactory. The optimal heat treatment regime should be performed on temperatures less than 550 °C, while cooling rate should be as slow as possible.

Although both investigated silane finishes have chemical similarity to the epoxy matrix: AMEO (containing an amino group, as does epoxy resin hardener) and GLYMO (containing an epoxy group) composite samples with

AMEO finish have, for all investigated heat-treatment regimes, from 13.5 to 14.5 % higher tensile strength than composites prepared with GLYMO finish.

In this work for the first time the possibility of using domestic glass fabric (TEHNOTEX, Sombor No. 558) for production of printed circuit boards for microelectronics has been confirmed. After appropriate heat treatment (500°C, for 60min, cooling rate 0.17°C/min) followed by chemical treatment with silanes having amino functional group like AMEO (Dynasilan® organo-functional silane) the investigated glass fabric together with epoxy matrix (resin Cy 233, HT 972 hardener and XB 3022 accelerator all from Ciba-Geigy) gives laminar composites with appropriate mechanical properties that satisfy the quality needed for production of printed circuit boards for microelectronics.

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