NASA TECH BRIEF

Lewis Research Center

TECHNIQUE FOR THE POLYMERIZATION OF MONOMERS FOR PPQ/GRAPHITE FIBER COMPOSITES

Because of processability problems, the potential of polyphenylquinoxalines (PPQ) as high temperature matrix resins in advanced composites has not been realized. Current methods used to fabricate PPQ/fiber composites consist of impregnating the reinforcement with high molecular weight PPQ polymer dissolved in m-cresol or a solvent mixture containing m-cresol. Because of the inordinately high viscosity of these solutions, which results from both the high molecular weight of the polymer and the use of m-cresol, complete wetting of the fiber during impregnation is difficult to achieve. The approaches used to overcome the viscosity problem are (1) to limit the polymer molecular weight by upsetting the stoichiometry of the system or (2) use dilute solutions. Upsetting the stoichiometry can adversely affect the polymer thermo-oxidative stability whereas dilute solutions of high molecular weight polymer are extremely viscous.

A novel approach has been developed to circumvent some of the composite processing problems associated with the use of PPQ polymers as the matrix material. The technique used to prepare high performance PPQ/graphite fiber composites consisted of impregnating the fiber with a freshly-made solution of the appropriate monomers instead of a solution of high molecular weight polymer.

Impregnation of the fiber prior to appreciable polymerization completely eliminates the impregnation problems encountered with the use of high viscosity high molecular weight PPQ solutions. It is important to note that the major part of the polymerization of the reactant mixture is conducted on the fiber during the solvent removal and final curing stages. Although at the time of impregnation the solution is not truly monomeric, this approach is referred to as the in situ polymerization of monomers because of its similarity to the monomeric reactant approach developed for A-type polyimides as described in NASA Tech Brief 71-10442.

In general, the following procedure was used to prepare a typical PPQ/graphite fiber composite. Stoichiometric quantities of bis(o-diamine) and bis(phenylglyoxal) were dissolved separately in N-methyl-pyrrolidone at a solids content of about 35 weight percent. The two solutions were then mixed and used to impregnate the graphite fiber as it was drum wound. Depending on the reactivity of the monomers, solvent, concentration, etc., the useful life (pot life) of the solution can range between one-half and six hours. For example, the solution of monomers for the typical polymer was used one-half hour after preparation with no difficulty. Polymerization and simultaneous solvent removal were caused to occur by heating the drum wound prepreg for 45 minutes at 344 K (160°F). Subsequently, the prepreg was removed from the drum, cut into plies 7.6 x 10.2 cm (3 x 4 inches), stacked (10 plies), wrapped in aluminum foil and placed into a press heated to the cure temperature, 602 K (625°F). Following a dwell time of about 90 seconds, pressures in the range of 1.38 to 6.89 MN/m^2 (200 to 1000 psi) were applied for one-half to one hour. In some instances, the composites were given elevated temperature post-cures, with and without applied pressure.

Some preliminary room temperature test results for this typical PPQ/graphite composite are as follows:

Flexural Strength	1289 MN/m^2 (187,000 psi)
Interlaminar Shear Strength	103 MN/m^2 (15,000 psi)
Transverse Tensile Strength	80 MN/m^2 (11,600 psi)
Longitudinal Impact Strength	3.05 joules (27 in-lb)

The higher flexural and interlaminar shear strengths can be attributed to the graphite fiber. The most significant point to be made is that the transverse tensile strength is approximately 50% higher than that for A-type polyimide/graphite composites. Also significant is the finding that the longitudinal impact strength of PPQ/graphite composites is 3 times that of epoxy/graphite composites.

(continued overleaf)

Notes:

1. This technique for the polymerization of monomers can be used to prepare high performance PPQ/graphite fiber composites, can greatly simplify the fabrication of PPQ fiber reinforced composites, and can eliminate the need for apriori polymer synthesis.
2. Additional information is contained in an article entitled "In Situ Polymerization of Monomers for High Performance PPQ/Graphite Fiber Composites" (to be published) in the JOURNAL OF APPLIED POLYMER SCIENCE.
3. No additional documentation is available. Specific questions, however, may be directed to:

   Technology Utilization Officer
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   Reference: B73-10014

Patent Status:

Inquiries concerning rights for the commercial use of this invention should be addressed to:

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