STRUCTURE-PROPERTY CORRELATIONS IN NEAT AND MICA-FILLED EPOXY RESIN

by

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CERTIFICATE

This is to certify that the thesis entitled "Structure-Property Correlations in Neat and Mica-filled Epoxy Resin", being submitted by Mr. C. Brahatheeswaran, to the Indian Institute of Technology, Delhi, for the award of the degree of Doctor of Philosophy in the Department of Textile Technology, is a record of bonafide research work carried out by him. Mr. C. Brahatheeswaran has worked under my guidance and supervision and fulfilled the requirements for the submission of the thesis.

The results contained in this thesis have not been submitted, in part or in full, to any other Institute or University for the award of a degree or diploma.

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ABSTRACT

The studies presented in this thesis relate to neat and mica-filled cured epoxy samples. The crosslinked networks of the neat resin samples were first characterized for molecular packing. Next the internal stress which was acquired by the unfilled and filled samples during the cooling part of the cure cycle was measured. The dynamic spectra of the various samples were then mapped in torsion and finally their stress-strain behaviour and electrical properties investigated. An attempt has been made to interpret the results of these investigations in a manner which is consistent with current thinking on these topics of scientific and technological interest.

The epoxy resin used was a diglycidyl ether of Bisphenol-A which was cured with m-phenylene diamine. Besides a stoichiometric composition of the epoxy resin and amine, off-stoichiometric samples containing excess epoxy and excess amine, were also prepared. Two curing cycles were used. The mica filler was of Muscovite type of Indian origin and was in the form of powder and flakes of two different sizes.

The volume expansion of the neat resin of different stoichiometries was measured from room temperature to about 200°C using mercury dilatometers. Combining the volume expansion data with the measured density values, specific volume-temperature plots were constructed and the empty and

free volume fractions determined. It was observed that the most highly crosslinked samples had the lowest measured density at room temperature. These highly crosslinked samples also had the highest free volume fraction and as a result, the intermolecular distance was high. This was attributed to the fact that the crosslink site does not provide a suitable environment for close packing of the rigid molecules.

The internal stress generated in the unfilled and filled samples during the cooling part of the cure cycle was estimated from the curvature acquired by a thin aluminium strip coated with the resin or mica-filled resin mixture. The relatively high internal stress generated in neat samples, subjected to high temperature post-cure treatment, was attributed to the greater amount of entrapped free volume. This internal stress was found to relax at a rapid rate, mainly as a result of molecular conformational changes. The internal stress in mica-filled samples relaxed almost completely through debonding of the mica-filler interface, delamination of the mica flakes and matrix cracking in addition to molecular conformational changes that occur in neat resin.

The dynamic studies carried out on a torsion pendulum gave interesting insight into the main relaxations shown by the neat resin in the unfilled samples and by the matrix resin in the filled samples. The differences between the two responses were analyzed and suitably explained. The

torsional modulus data of the various samples was discussed in terms of the stiffening effect of the mica filler which results in constraints on mobility. The α'-relaxation was shown to correlate well with the internal stress developed in the sample. The glass transition temperature (T_g), as determined from the α-relaxation peak, was compared with T_g obtained from dilatometry and differential scanning calorimetry. The tensile stress-strain studies showed that while the moduli of the filled samples were higher than that of the neat cured resin, their strength and elongation-to-break were considerably reduced. The constraints to resin deformation due to the presence of the filler, the existence of internal stress and the cracks in the matrix were considered to be the important factors affecting the tensile mechanical properties. An analysis of the data in terms of simple existing models has also been made.

The measurement of dielectric constant and loss factor over a range of temperature and frequency showed that network characteristics like molecular packing and internal stress and the constraints to molecular mobility in the presence of mica affected the electrical properties also.