

**MECHANICAL PROPERTIES OF RUBBERY
COPOLYMER GRAFTED SILICA FILLED EPOXY
NANOCOMPOSITES AND INTERFACE STUDY**

By

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ABSTRACT

Epoxies are widely used as an important insulation component in high voltage rotating machines due to their good mechanical strength, excellent electrical insulation ability, good thermal stability, and easy processability. However, epoxies are brittle and have poor fracture toughness, which can lead to catastrophic failures where they are used. Rubber or rigid particles are often added to toughen epoxies. Rubber particles improve ductility, but decrease the mechanical strength of epoxy matrices. Rigid particles, on the other hand, decrease ductility but increase mechanical strength. Improvement in both properties can be achieved by using a combination of rubber and rigid fillers. However, a high loading of fillers is required, making processing difficult.

In polymer nanocomposites, the surface area to volume ratio of the nanoparticle is very large, which means the interphase properties are essential in determining the mechanical properties of the nanocomposite. A strong interphase often leads to enhancement in strength, while a softer interphase can contribute to improving composite ductility. Ideally, the interphase properties can be tailored to achieve desired nanocomposite properties. However, the effects of interphase are not well understood and modifying the interphase at will using conventional silane treatments is also challenging.

In this work, copolymers were grafted to SiO_2 nanoparticles to form a controllable interphase to improve the fracture toughness and ductility of an epoxy without decreasing its modulus. The copolymer was composed of a rubbery inner block (poly(hexyl methacrylate), PHMA) and an epoxy compatible outer block (poly(glycidyl methacrylate), PGMA). Copolymers with graft densities from 0.07 to 0.7 chains/ nm^2 and molecular weights of PHMA from 20 to 80 kg/mol were synthesized in order to generate a wide range of interphase mechanical properties. The PHMA inner block thickness, measured by AFM, ranged from 4 to 18 nm, and increased with increasing graft density and molecular weight. The elastic modulus of the rubbery inner block increased with decreasing molecular weight and increasing

graft density. The tensile strength decreased proportionally with PHMA concentration for all the copolymer grafted SiO₂ filled nanocomposites, due to the low strength of the PHMA inner block.

It was found that PHMA polymer chains with low graft density and high molecular weight achieved the best overall mechanical properties. The ductility of the nanocomposites (as measured by the tensile strain-to-failure) was 35 % better than the neat epoxy for a 0.07 chains/nm² PHMA block with a molecular weight of 80 kg/mol at 1.5 vol % of the SiO₂ core. The fracture toughness (K_{1C}) was three times that of the neat epoxy and the fatigue crack growth rate was reduced by two orders of magnitude at maximum. A 10 % increase in Young's modulus was also seen in this nanocomposite.

The leading toughening mechanisms in the copolymer grafted SiO₂ filled epoxy nanocomposites were found to be plastic void growth and shear banding. Unlike conventional rubber particles, which cavitate to release external hydrostatic stress, the rubbery interphase did not fail but stretched to release the hydrostatic stress. A polymer physics model was used to study the deformation behavior of the rubbery inner block. It was found that at low graft density and large molecular weight, the rubbery interphase can achieve larger deformation than that of a rubber particle before cavitation, allowing the rubbery interphase to toughen the epoxy at filler sizes that conventional rubbers cannot toughen the epoxy.