

IMPACT OF PREPARATION METHODS ON DIELECTRIC AND MECHANICAL PROPERTIES OF GRAPHENE-EPOXY NANOCOMPOSITES

Ziani S.B.de Souza^{1,2}, Guilhermino J. M. Fechine^{3,4}, Maurício A. da Motta Sobrinho², Éric David¹, Nicole R. Demarquette^{1*}

¹Department of Mechanical Engineering, École de Technologie Supérieure, Montreal, Canada

²Department of Chemical Engineering, University Federal of Pernambuco, Recife, Brazil

³School of Engineering, Mackenzie Presbyterian, São Paulo, Brazil

⁴Mackenzie Institute of Research in Graphene and Nanotechnologies - MackGraphe, Mackenzie Presbyterian Institute, São Paulo, Brazil

* nicoler.demarquette@etsmtl.ca

ABSTRACT

Proper dispersion of graphene in epoxy is essential for enhancing nanocomposite properties and requires control of processing methods. Here, epoxy/graphene composites (E/G, 1% wt) were prepared via solution mixing. To evaluate the effect of additional dispersion treatments, the graphene/acetone suspensions before mixing with epoxy, or uncured E/G samples, were further mixed using an ultrasonic bath (UB) or ultrasonic probe (UP). The rheological properties of the uncured materials were analyzed performing small amplitude oscillatory shear (SAOS). The morphology of the cured samples was observed by optical microscopy and their electrical and mechanical properties by broadband dielectric spectroscopy, and dynamic mechanical analysis (DMA) respectively.

The modulus of the composites varied depending on the treatment applied. Untreated E/G exhibited a lower storage modulus (G') at low frequencies compared to the neat epoxy. This is attributed to the slippage of aggregates within the composite, a phenomenon known as "lubrication effect." UB treatment of the E/G composite did not significantly impact G' . Conversely, applying UB (1 h) to the graphene/acetone suspension increased G' , indicating an improved graphene distribution, favored by processing at lower viscosity that may facilitates the ultrasonic wave propagation. Microscopy images confirmed reduced agglomerates, though average particle size remained unchanged.

UP treatment was more effective, leading to a higher G' after 10 minutes at 25% amplitude. However, excessive UP treatment (75% amplitude for 30 minutes) reduced G' . The lower G' is associated with graphene sheet fragmentation, as observed in microscopy images showing reduced sheet size, potentially compromising reinforcement. Direct UP application to the E/G composite led to polymer degradation.

The dielectric properties of neat epoxy and E/G composites were analyzed using a combination of Havriliak-Negami function and a conductivity term to take into account charge fluctuations. The relaxation time of the dipolar processes was further modeling with the Vogel-Fulcher-Tammann equation providing the activation energy of the relaxation mechanisms, reflecting molecular mobility restrictions. While untreated E/G showed activation energy similar to that of neat epoxy, UB treatment increased activation energy, suggesting improved graphene dispersion and greater restriction of molecular motion of the epoxy chains. This was confirmed by DMA, which showed increased storage modulus, loss modulus, and glass transition temperature, indicating a more rigid polymer with enhanced filler-matrix interactions.

In conclusion, this study highlights the importance of optimizing processing parameters. The UB applied to the graphene suspension improved the dispersion without fragmenting the sheets, resulting in higher activation energy and improved dynamic mechanical properties.