

Interfacial Polarization in Polymer-Based Dielectric Composites with 2D Nanomaterials

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The dielectric behavior of polymer-based composite films was systematically investigated with a particular focus on the interfacial polarization phenomena arising from the incorporation of conducting two-dimensional (2D) nanoplatelets. Ferroelectric BaTiO₃ (BTO) nanoparticles were employed as a primary filler; while MXene, graphene oxide (GO), and reduced GO (rGO) served as co-fillers to modulate the interfacial effects. Cyanoethyl pullulan (CEP) was used as a polymer matrix due to its high dielectric behavior among polymers. A reference sample, containing BTO nanoparticle fillers only in a CEP matrix, was also prepared to isolate the contribution of the 2D nanomaterials, which resulted in four dielectric composite samples. MXene platelets were delaminated in dimethyl sulfoxide (DMSO) via ultrasonication for 2 hours, and rGO was chemically reduced from GO using hydrazine over 8 hours—both conditions optimized in prior studies [1,2] to enhance dielectric performance. All components were dispersed in dimethylformamide (DMF) and spin-coated onto ITO substrates to form uniform dielectric films.

The incorporation of conducting 2D nanoplatelets significantly enhanced the dielectric constant of the composites. The highest value was achieved with the MXene-incorporated film (~200 at 1 kHz), followed by GO films (~130) and rGO films (~120), all substantially higher than the reference film containing BTO only (~90). However, this enhancement was accompanied by an undesirable increase in the dielectric loss ($\tan \delta$) due to percolative linkage of fillers. The loss increased, from 0.048 for the reference sample, to 0.053 for GO, 0.10 for MXene, and 0.22 for rGO, respectively. The observed dielectric enhancement is attributed to pronounced interfacial polarization at the filler-matrix boundaries, facilitated by the high aspect ratio and conductivity of the 2D nanoplatelets. Among them, MXene demonstrated superior interfacial coupling due to its metallic conductivity, leading to more effective charge accumulation at interfaces. In addition, MXene can successfully suppress the increase in the dielectric loss which is difficult to control for nanomaterial incorporation within a polymer matrix. Raman spectroscopy and X-ray photoemission spectroscopy (XPS) analyses corroborated the structural and electronic characteristics responsible for these effects. These findings demonstrate the critical role of interfacial polarization in tailoring the dielectric properties of polymer-based composites and suggest that MXene-based systems hold promise for next-generation electronic and energy storage applications, where high permittivity and controlled loss are essential.

[1] S.-Y. Jun and S. Yu, J. Korean Phys. Soc. **82**, 1093 (2023).

[2] S.-Y. Jun. *et al*, Carbon **199**, 23 (2022).

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Supplementary Pages (Optional)

The following two figures are observed dielectric characteristics for four samples: the reference sample with BTO only, GO-incorporated one, rGO-incorporated one, and MXene-incorporated one. The right figure is the diagram for the values of dielectric constant and dielectric loss measured at 1 kHz, to figure out the trade-off relation between dielectric constant and dielectric loss comprehensively. We can say that MXene is the best choice as a 2D nanomaterial filler for polymer-based dielectric composites.

