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Effect of Filler Concentration and Frequency on the Dielectric Properties of Bitter Cola Seed Carbonized HDPE

UCHECHUKWU VINCENT OKPALA[1], ODUNZE CHIKE BARTHLOMEW[2], JOY IFEOMA EMEODI[3]
[1,3] *Departments of Industrial Physics, Chukwuemeka Odumegwu Ojukwu University, Anambra State, Nigeria.*
[2]*Department of Physics/electronics, School of Applied Sciences, Imo State Polytechnic, Omuma, Imo State, Nigeria*

Abstract- ***In this work, we studied the development of a sustainable, high-performance green dielectric material. This was achieved through the use of bitter cola (Garcinia kola) seed as a conductive filler in a High-Density Polyethylene (HDPE). The bitter cola seeds were carbonized at 700°C under an inert atmosphere to get biochar. It was mixed and compressed with melted high density polyethylene. This enabled us to form composite sheets with varying filler concentrations of 0 wt.%, 5 wt.%, 10 wt.%, 15 wt.%, and 20 wt.%. We determined the dielectric properties; relative permittivity (ε′), dielectric loss factor (ε″), and loss tangent (tanδ) over a wide frequency spectrum of 102 Hz to 106 Hz at room temperature. The results showed that the relative permittivity increased non-linearly with biochar loading, thereby showing a dramatic enhancement, as the concentration approached the percolation threshold at approximately 15 wt.%. Conversely, a strong frequency dispersion was observed as ε′ and ε″ decreased significantly with increasing frequency. This behavior was accurately described by the Maxwell-Wagner-Sillars (MWS) interfacial polarization mechanism. This work introduced a viable, low-cost path for green functional bio-composites for embedded capacitors, and electromagnetic interference (EMI) shielding applications.***

I. INTRODUCTION

The relentless miniaturization and increasing speed of modern microelectronic devices have intensified. The demand for advanced polymer dielectric materials with tunable properties, is a function of increased miniaturization and speed of modern microelectronics (Wang et al., 2023). The traditional inorganic ceramics offer high dielectric constants, but their brittle nature, high density, and difficulty in processing present a serious engineering challenges. This is why polymer matrix composites (PMCs) reinforced with functional fillers offer a compelling alternative. Its flexibility, ease of processing, and high breakdown strength with host polymers, gives the fillers superior electrical or dielectric features, (Deeba et al., 2022).

We chose High-Density Polyethylene (HDPE) due to its exceptional chemical resistance, good mechanical properties, low cost, and inherently low dielectric loss of tan $\delta = 10-4$. However, its clean non-polar structure yields a low relative permittivity ε' of 2.3, thereby limiting its direct performance in energy storage applications. According to literature, the dielectric performance of polymers, carbonaceous nanofillers like graphene, carbon nanotubes (CNTs), and carbon black have been extensively investigated (Bartoli et al., 2022; Putson et al., 2011). In spite of these facts, their high production costs and tedious processing methods limit mass adoption, sparking a shift toward sustainable alternatives.

Biochar (BC) from the pyrolytic carbonization of agricultural waste at a temperature of about 600oC to 800oC is a formidable alternative to synthetic carbonaceous materials. Processing bitter cola seed into functional carbonaceous fillers, puts it into advanced engineering applications.

This is why in this paper, we evaluated the combined effects of filler concentration, and applied electric field frequency on the complex dielectric properties of carbonized Bitter Cola seed/HDPE composites. We systematically elucidate the polarization mechanisms governing the low-frequency and high-frequency boundaries to provide structural insights for tailoring sustainable green electronics.

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II. EXPERIMENTAL METHODOLOGY

2.1 Materials

In this work commercial injection-grade High-Density Polyethylene (HDPE) of density: 0.954 g/cm3, melt flow index: 8.0 g/10 min was used as the polymer matrix. Garcinia kola seed bought from different markets were dried in open air for one month. They were washed with ordinary water, and deionized water to eliminate pulp residues. They were later oven dried at 105∘C for 24 hours to remove bound moisture according to Yim & Kim, 2023. The dried biomass was placed inside a horizontal quartz tube furnace and carbonized at 700∘C for 2 hours, at a heating rate of 10∘C/min. The carbonized residue was ball-milled at 400 rpm for 4 hours, and sieved to yield fine micro-particles of 32 μm.

2.1 Fabricating the Composites

The composites were prepared using two different step processes:

1. Melt Compounding: Here, HDPE pellets and carbonized biochar powders dried at 80∘C were processed, using a twin-screw extruder at 180∘C and screw speed of 60 rpm. The fillers were loaded at 0 wt.%, 5 wt.%, 10 wt.%, 15 wt.%, and 20 wt.%.
2. Compression Molding: The extruded strands were pelletized and hot-pressed at 190∘C under the pressure of 15 MPa for 8 minutes, to achieve uniform square sheets.

2.3 Dielectric Measurements

Circular aluminum electrodes of 20 mm in diameter were sputter-coated onto both faces of the disc specimens, to guarantee ideal electrical contact. High-precision LCR Impedance Analyzer was used to record the relative permittivity (ε′), dielectric loss factor (ε′′), and loss tangent (tanδ), across a frequency range of 102 Hz to 106 Hz at room temperature.

III. RESULTS AND DISCUSSION

3.1 Effect of Filler Loading on Dielectric Permittivity

On table 1.1 are the changes in relative permittivity (ε′) at different frequencies against filler loading.

Table 1.1: Dielectric parameters of Bitter Cola BC/HDPE composites at specified frequencies.

Filler Loading (wt. %)	ε^1 at 10^2 Hz	ε^1 at 10^4 Hz	ε^1 at 10^6 Hz
0% (Pure HDPE)	2.31	2.30	2.29
5% BC	5.45	4.80	4.12
10% BC	12.80	9.95	7.30
15% BC	48.60	28.10	14.50
20% BC	185.20	65.40	22.10

Table 1.1. shows that the real permittivity (ε′) increased across all frequencies, as carbonized biochar were increased. The plot of real permittivity ε′ against filler loading (wt.%) is as shown on figure 1.1.

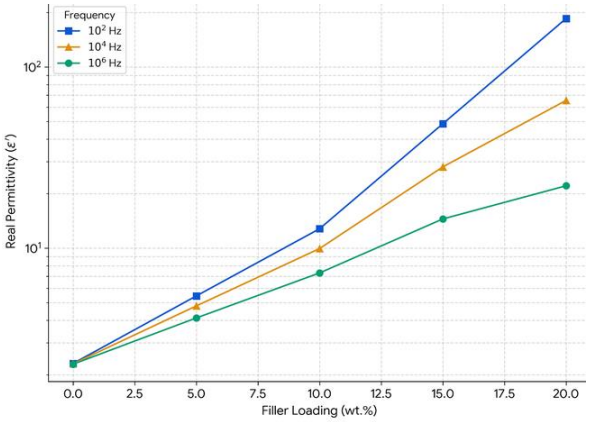

Figure 1.1: Real permittivity ε′ versus filler loading (wt.%).

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This was in agreement with two complementary factors:

1. The Dipolar/Interfacial Polarization Mechanism: The introduction of semi-conductive carbon with structural defects into a non-polar polymer created an extreme mismatch, in electrical conductivity (σ) and permittivity (ε) across the internal phase boundaries (Bafna et al., 2023; Wang et al., 2023).
2. Maxwell-Wagner-Sillars (MWS) Effect: This posits that free migrating charge carriers rapidly accumulate at the highly disordered matrix-filler interfaces, under an external field (Bafna et al., 2023). These trapped charges form massive macro-dipoles, generating a strong internal space-charge polarization that yields high capacitance values (Bafna et al., 2023).

The sharp surge in ε′ near 15 wt.% indicated that the composite was approaching its structural percolation threshold (fc). This was modeled by the power law equation (Putson et al., 2011):

$$\varepsilon' = \varepsilon_m^1 \left| \frac{f_c - f}{f_c} \right|^{-q} \quad for\ f < f_c$$

where ε_m^1 represents the permittivity of the host matrix, f represents the filler volume fraction, and q represents a critical scaling exponent (Putson et al., 2011). At 20 wt.%, the particles surpassed this threshold, allowing charges to tunnel across adjacent conductive pathways and causing ε′ to climb significantly to 185.20 at 102 Hz.

3.2 Frequency-Dependent Dielectric Behavior

The frequency dispersion curves reveal a distinct behavioral shift across different operational windows:

- Low-Frequency Regime (102 Hz−103 Hz): The composites demonstrated high permittivity values. At these frequencies, the alternation of the electric field was slow enough to allow migrating electrons sufficient relaxation time to migrate and accumulate at the BC-HDPE boundary layer interfaces (Bafna et al., 2023; University of Babylon Private CDN).
- High-Frequency Regime (104 Hz−106 Hz): ε′ dropped sharply, thereby converging toward a baseline value. This drop occurred because the rapid field reversals outpaced the relaxation timescales of the space charges (University of Babylon Private CDN). The dipoles simply cannot polarize quickly enough to track the field oscillations. This caused the MWS interfacial polarization to collapse thereby leaving only rapid, low-loss electronic or atomic polarizations active (Bafna et al., 2023; University of Babylon Private CDN).

3.3 Dielectric Loss Factor (ε″) and Loss Tangent (tanδ)

The loss tangent (tanδ = ε″/ε′) acts as a direct measure of energy dissipation within the system (Ellison et al., 2017; Wang et al., 2023). As expected, tanδ increased alongside filler concentrations due to elevated conduction losses. Below the percolation limit (≤10 wt.%) the loss tangent remained remarkably low (tanδ < 0.05 at 102 Hz), revealing that the isolated biochar particles primarily acted as micro-capacitors without triggering leakage currents.

However, at 20 wt.% filler loading, tanδ scaled past unity (tanδ = 1.15). This behavior occurs because the fillers formed continuous conducting networks, shifting the energy dissipation mechanism from mild polarization relaxation to active ohmic conduction paths, where leakage currents dominated (Bafna et al., 2023).

IV. CONCLUSION

Sustainable green dielectric composites were successfully made, by incorporating carbonized bitter cola (Garcinia kola) seed biochar into an HDPE matrix. The real permittivity (ε′) increased efficiently with filler loading, reaching a high value of 185.20 at 102 Hz for the 20 wt.% composite, compared to just 2.31 for pure HDPE. The frequency dependence of ε′ and tanδ verified that Maxwell-Wagner-Sillars interfacial polarization governs the material's low-frequency performance. Finally, choosing a low filler loading of 10 wt.%, engineers can achieve a fivefold increase in permittivity, while still maintaining low dielectric loss. This will make the composites good for flexible, and sustainable electronic components.

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