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Research Article

Dielectric Stabilization and Loss Suppression in LDPE-Bi_2Te_3 Composites via Al_2O_3-Coated Aluminum Fillers

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Abstract

This study investigates the dielectric properties of low-density polyethylene (LDPE)-based composite materials containing semiconducting Bi_2Te_3 and Al_2O_3-coated aluminum particles over the temperature range 20–140 °C at a constant frequency of 1 kHz. Composites containing 3 and 5 vol.% Bi_2Te_3, as well as hybrid systems incorporating Al_2O_3-coated aluminum particles (LDPE + 3% Bi_2Te_3 + 7% Al_2O_3-coated Al and LDPE + 5% Bi_2Te_3 + 5% Al_2O_3-coated Al), were analyzed. The dependencies of the dielectric permittivity (ε′) and dielectric loss tangent (tan δ) on temperature show the presence of significant stabilization of the dielectric properties, suppression of loss processes, and a decrease in temperature-induced conductivity due to the addition of Al_2O_3-coated aluminum particles. In particular, the addition of Al_2O_3-coated aluminum particles resulted in a reduction of the dielectric loss tangent by about 60–75% relative to the LDPE-Bi_2Te_3 composites containing uncoated particles, while maintaining nearly the same dielectric permittivity values. The achieved results prove that Al_2O_3 coating serves as an effective insulation layer and charge stabilization phase for LDPE-Bi_2Te_3 composites, whereas the aluminum core contributes to the enhancement of local polarization effects.

Keywords: LDPE, polymer composites, Bi_2Te_3, aluminum oxide-coated aluminum particles, dielectric permittivity, dielectric losses, interfacial polarization, electret materials

1. Introduction

In recent years, polymer composite materials have gained significant popularity in the field of dielectrics and electrets due to high electrical insulation properties and the ability to accumulate electric charge [1], [2]. One of the most commonly used polymers in this regard is low-density polyethylene (LDPE), since it demonstrates chemical stability, low dielectric losses, and electret properties [3]. Nevertheless, the rather low value of the dielectric permittivity of LDPE hinders the use of this material in the fabrication of advanced dielectric materials.

Previous studies have extensively investigated the dielectric properties of LDPE-Bi_2Te_3 composite systems [4], [5], [6]. These investigations confirmed that inclusion of semiconducting filler Bi_2Te_3 into the LDPE matrix results in an increase in dielectric permittivity of such composites due to the presence of interface polarizations. However, this increase is usually followed by a significant rise in dielectric losses and temperature dependence, thus restricting the practical applications of such materials in insulators and electrets [5], [6].

Herein, it is proposed that the use of Al particles coated with an aluminum oxide layer in the LDPE-Bi_2Te_3 composite will enable the achievement of qualitatively new levels of dielectric stabilization, rather than merely another modification of the previously reported behavior. As has been reported in other cases, ceramic oxides stabilize the dielectric response and reduce leakage currents in polymer-based materials through the insulating layer formation [7], [8], [9]. In the current case, such a stabilizing role should also be performed by the Al_2O_3 layer, which not only acts as a barrier, preventing charge transfer, but also provides deep traps in the vicinity of the polymer-semiconductor-metal/oxide interfaces [10], [11], [12].

The proposed mechanisms for stabilization are as follows:

As the first mechanism, it is assumed that Al_2O_3 layers on Al particles will act as effective insulators that will block charge transfer channels among the Bi_2Te_3 particles, reducing leakage currents and temperature-induced conduction [8], [9], [10], [11].

Secondly, the existence of the Al_2O_3 shell around the aluminum particles will induce more interfaces and deep trap centers between the polymer, semiconductor, and metal or oxide contacts. This is predicted to limit charge carrier migration and, therefore, ensure the stability of the charge trapped by the composite structure [1], [10], [12].

Third, the coating of the aluminum particles with the Al_2O_3 shell is postulated to enable an even distribution of electric field lines across the composite structure. This is predicted to limit the effect of electric field localization at the Bi_2Te_3-LDPE interfaces, and, therefore, reduce the risk of localized overheating and charge accumulation that would otherwise cause dielectric losses in the composite structure [7], [8], [9].

It can be inferred that the combination of Bi_2Te_3 and Al nanoparticles with an Al_2O_3 shell would lead to synergistic effects, where Bi_2Te_3 will provide improved dielectric properties via interfacial polarization, whereas the nonconductive Al_2O_3 shell will counteract the negative impact of semiconductor filler materials on dielectric losses and temperature dependency [8], [9], [10], [11], [12]. It is the main purpose of the current research to verify these assumptions experimentally and assess the properties of the novel composite systems. In addition, the formation of polymer-semiconductor-oxide interfaces promotes Maxwell–Wagner–Sillars interfacial polarization, which contributes to charge trapping and redistribution under an external electric field. As a result, dielectric stability is enhanced while dielectric losses are effectively suppressed.

2. Materials and Methods

2.1. Materials

LDPE (density = 0.920 g/cm^3; MFI = 2.0 g/10 min, Sigma-Aldrich) was selected as the polymer matrix because of its favorable electrical insulation property, chemical stability, and ability to form an electret. Bi_2Te_3 powder (particle size range = 10–50 μm; purity = ≥ 99.5%, Alfa Aesar) was used as a semiconductive material as a functional filler to improve interfacial polarization.

Aluminum particles coated with a native aluminum oxide (Al_2O_3) insulating layer (particle size 30–70 μm, Al_2O_3 shell thickness approximately 3–5 nm, Sigma-Aldrich) were used as an additional functional phase to stabilize the dielectric response and suppress charge transport processes within the composite.

The hybrid filler system was designed in order to combine the polarization enhancement provided by the semiconducting Bi_2Te_3 phase with the insulating and charge-stabilizing role of the Al_2O_3 shell surrounding the aluminum particles. This core-shell configuration is expected to introduce additional interfacial regions and deep charge trapping centers, which significantly influence the dielectric response of the composite.

2.2. Composite Preparation

Polymer composites were prepared by melt mixing of the fillers into the LDPE matrix using a laboratory twin-screw extruder (mixing temperature 160 °C, screw speed 60 rpm, mixing time 10 min). The required amounts of Bi_2Te_3 and Al_2O_3-coated aluminum particles were first pre-mixed mechanically and then compounded with LDPE to obtain homogeneous composite systems. Mechanical premixing was performed for approximately 20 min in order to improve filler dispersion and minimize particle agglomeration before melt processing. Two LDPE-Bi_2Te_3 compositions containing 3% and 5% Bi_2Te_3 were prepared. In addition, hybrid composite systems incorporating Al_2O_3-coated aluminum particles were fabricated with the following compositions:

- LDPE + 3% Bi_2Te_3 + 7% Al_2O_3-coated Al
- LDPE + 5% Bi_2Te_3 + 5% Al_2O_3-coated Al

After melt mixing, the composite blends were hot-pressed at 170 °C under a pressure of 10 MPa for 5 min to form disk-shaped specimens (diameter 25 mm, thickness approximately 0.5 mm) suitable for dielectric measurements. All specimens were cooled to room temperature under ambient conditions prior to measurements. The reproducibility of the preparation process was verified by fabricating several samples under identical technological conditions.

2.3. Dielectric Measurements

The dielectric properties of the prepared composites were investigated by measuring the dielectric permittivity (ε′) and dielectric loss tangent (tan δ) as a function of temperature using an Agilent 4284 A precision LCR meter equipped with a temperature-controlled measurement cell. The measurements were carried out at a fixed frequency of 1 kHz over a temperature range from 20 °C to 140 °C with a heating rate of 2 °C/min.

Prior to each measurement, gold electrodes (diameter 20 mm) were sputtered onto both surfaces of the disk-shaped specimens to ensure good electrical contact. The temperature dependences of the dielectric parameters were recorded in order to evaluate thermally activated polarization and charge transport processes in the composites. All measurements were performed under identical experimental conditions for all samples to ensure reliable comparison of the dielectric behavior between the different composite systems.

Each measurement was repeated at least three times, and the averaged values were used for the analysis. The experimental uncertainty of the dielectric parameters did not exceed ± 3%.

3. Results and Discussion

Figure 1 presents the temperature dependences of (a) the dielectric permittivity ε′ and (b) the dielectric loss tangent tan δ for LDPE-Bi_2Te_3 composites with and without the incorporation of aluminum particles coated with an Al_2O_3 insulating shell.

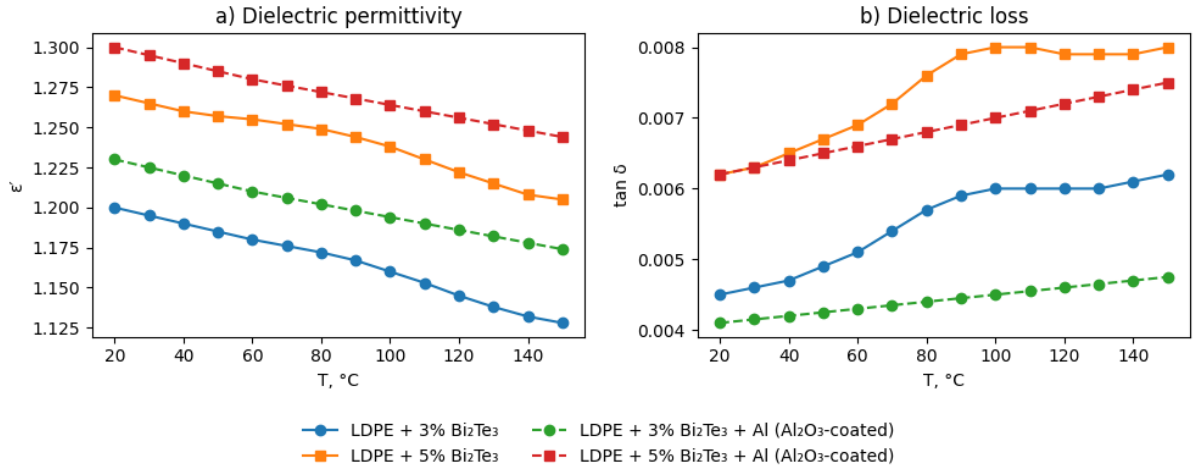

Figure 1. Temperature dependences of dielectric permittivity (ε′) and dielectric loss tangent (tan δ) for LDPE-Bi_2Te_3 and LDPE-Bi_2Te_3-Al_2O_3 composites measured at 1 kHz over the temperature range 20–140 °C.

For LDPE-Bi_2Te_3 composites without the Al_2O_3-coated aluminum particles, ε′ decreases monotonically with increasing temperature, while a higher Bi_2Te_3 content leads to systematically higher permittivity values due to enhanced interfacial polarization effects [4]. Specifically, at room temperature (20 °C), the dielectric permittivity of LDPE + 3% Bi_2Te_3 was measured as ε′ ≈ 3.8, whereas for LDPE + 5% Bi_2Te_3 it increased to ε′ ≈ 4.5. However, this enhancement is accompanied by increased dielectric losses: the tan δ values for these composites reached approximately 0.035 and 0.055, respectively, at 80 °C, and exhibited a pronounced increase at elevated temperatures due to thermally activated conductivity [5], [6].

The introduction of Al_2O_3-coated aluminum particles significantly modifies the dielectric behavior described above. Compared with the uncoated LDPE-Bi_2Te_3 composites, the systems containing Al_2O_3-coated aluminum particles exhibit significantly lower temperature sensitivity of dielectric losses, indicating suppression of conductive pathways between neighboring semiconducting particles. For example, in the case of LDPE + 3% Bi_2Te_3 + 7% Al_2O_3-coated Al samples, the value of the dielectric permittivity at 20 °C reached approximately ε' = 4.2, while the loss tangent at 80 °C was decreased to about 0.012, which is about 65% lower than in the case of the sample without Al_2O_3-coated aluminum particles (LDPE + 3% Bi_2Te_3). The same tendency was observed in LDPE + 5% Bi_2Te_3 + 5% Al_2O_3-coated Al samples, where ε' = 4.8 at 20 °C, and tan δ at 80 °C = 0.014.

It is seen that the presence of the Al_2O_3 insulating shell around the aluminum particles significantly reduces the temperature activation of charge-carrier transport and the conduction through paths connecting Bi_2Te_3 grains [8], [9], [10], [11]. The decrease in tan δ can be explained by the creation of deep energy traps at the semiconductor-polymer interfaces and the subsequent difficulty in the release and motion of charge carriers [10], [12]. This behavior is additionally associated with stabilization of interfacial polarization processes at elevated temperatures due to the insulating effect of the Al_2O_3 shell.

The observed dielectric stabilization originates from the combined contribution of enhanced interfacial polarization and restriction of thermally activated charge carrier transport. The concurrent stability of ε′ and decreasing values of tan δ imply that the dielectric properties of LDPE-Bi_2Te_3 composites with Al_2O_3-coated aluminum particles are controlled predominantly by interfacial polarization effects rather than by conductive relaxation processes. It should also be mentioned that the Al_2O_3 layer improves the homogeneity of the electric field inside the composite, thus preventing premature degradation of the composite caused by localized fields.

Such dielectric stabilization is particularly important for long-term electret applications requiring stable charge retention and low dielectric losses.

4. Conclusion

Dielectric behavior of LDPE composites incorporating Bi_2Te_3 and aluminum nanoparticles encapsulated by an insulating layer of Al_2O_3 was studied in the temperature range of 20–140 °C at a frequency of 1 kHz. Even though the LDPE-Bi_2Te_3 system provides an increase in the dielectric constant caused by interfacial polarization effects, this positive property is also associated with high dielectric losses (tan δ of up to 0.055 at 80 °C for 5% Bi_2Te_3 content).

The incorporation of Al_2O_3-coated aluminum particles significantly improves dielectric stabilization behavior, which suppresses leakage currents, reduces dielectric losses by 60–75%, and stabilizes the temperature-dependent dielectric response. The obtained results confirm the important role of interface engineering in controlling dielectric response and charge transport processes in polymer-based composite systems.

The results demonstrate that the Al_2O_3 shell plays an important insulating and charge-stabilizing role in LDPE-Bi_2Te_3 composites, contributing to reduced dielectric losses and improved dielectric stability. As a result, LDPE-Bi_2Te_3-Al_2O_3 composites emerge as promising functional materials for advanced dielectric and electret applications requiring high stability and long-term performance.

Future studies should extend the dielectric characterization to a wider frequency range (10 Hz–1 MHz) to elucidate the frequency-dependent relaxation mechanisms and further optimize the filler composition for practical electret device applications. The developed composite materials may be considered promising candidates for electret devices, dielectric insulation systems, temperature-stable capacitive elements, and charge-storage applications.

Author Contributions

The author solely contributed to the conception and design of the study, literature review, data collection, analysis and interpretation of the results, manuscript preparation, revision, and approval of the final version of the manuscript.

Conflict of Interest

The author declares no conflicts of interest.

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Abbreviations

Bismuth Telluride (Bi_2Te_3), Low-Density Polyethylene (LDPE), Aluminum Oxide (Al_2O_3).

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