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Enhanced dielectric properties of HDPE nanocomposites incorporating HFO_2 and Ta_2O_5 nanoparticles

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Abstract

This study explores the structural and dielectric properties of high-density polyethylene (HDPE) nanocomposites incorporating HfO_2 and Ta_2O_5 nanoparticles before and after high-voltage corona discharge (CD) treatment. The CD process, involving charge transfer in a non-uniform electric field, was employed to evaluate surface modification and dielectric stability. Scanning Electron Microscopy (SEM) and Energy-Dispersive X-ray (EDX) analyses confirmed uniform nanoparticle dispersion within the HDPE matrix, with limited agglomeration at higher filler contents. The dielectric constant (ε′), measured as a function of frequency and nanoparticle concentration, increased with filler loading due to enhanced interfacial polarization, in good agreement with the Maxwell-Garnett theoretical model. After CD treatment, a minor reduction in permittivity was observed, likely caused by structural rearrangements and changes in charge-trapping dynamics. Overall, the results indicate that corona discharge modifies the interfacial polarization and stabilizes the dielectric response of HDPE/oxide nanocomposites, making them promising candidates for advanced high-voltage insulation applications.

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1. Introduction

Polymer-based nanocomposites have emerged as promising materials for advanced electrical and electronic applications due to their lightweight nature, processability, flexibility, and tunable dielectric performance [1-4]. Among them, high-density polyethylene (HDPE) is one of the most widely used insulating polymers because of its excellent mechanical strength, low dielectric loss, and high breakdown resistance [5]. However, its inherent limitations-such as relatively low dielectric permittivity and susceptibility to electrical ageing-have motivated extensive research on modifying its structure through the incorporation of high-permittivity oxide nanoparticles [6, 7, 8].

The introduction of metal oxide nanoparticles such as TiO_2, HfO_2, and Ta_2O_5 into polymer matrices significantly enhances their electrical, thermal, and interfacial polarization characteristics [9-14]. These fillers improve charge trapping, interfacial polarization, and dielectric stability under high-voltage stress conditions. For instance, Rahimli et al. [2, 3, 14-20] demonstrated that the inclusion of ZnO and TiO_2 nanoparticles into polystyrene matrices led to enhanced dielectric strength, improved charge transport regulation, and defect-related luminescence control. Similarly, Ramazanov et al. [6, 9, 10] reported that nanoparticle-induced interfacial polarization strongly influences the dielectric and photoluminescent properties of polymer nanocomposites such as PVC/TiO_2, PP/TiO_2, and PVDF/ZrO_2. These findings confirm that interfacial effects play a dominant role in governing the electrical response of polymer–oxide systems.

In recent years, hafnium dioxide (HfO_2) and tantalum pentoxide (Ta_2O_5) have attracted increasing attention as dielectric fillers due to their high permittivity, wide bandgap, and exceptional chemical stability [12]. When dispersed within polymer matrices, these oxides not only enhance the dielectric constant but also improve breakdown strength and energy storage density. Their strong ability to trap charge carriers helps suppress conduction losses, making them suitable for high-voltage insulation and energy storage applications [9, 12]. Nevertheless, the interfacial structure between the polymer and nanofiller, as well as the effects of electrical ageing, remain key factors determining overall dielectric performance.

Corona discharge (CD) is a powerful technique to investigate and modify the electrical stability of insulating materials [11]. It involves ionization in a high-voltage field, where energetic ions interact with the polymer surface, altering charge distribution, trap density, and interfacial states. Previous studies have shown that CD exposure can induce oxidation, chain scission, and reorganization of the polymer surface, which in turn affect dielectric and conductive properties [11, 12]. Understanding these changes is essential for optimizing the reliability of polymer nanocomposites used in high-voltage environments.

In this work, HDPE-based nanocomposites containing varying concentrations of HfO_2 and Ta_2O_5 nanoparticles were prepared and subjected to corona discharge treatment. The study aims to elucidate the relationship between nanoparticle concentration, interfacial polarization, and the dielectric response before and after CD. Structural and morphological analyses (SEM/EDX) were performed to verify nanoparticle dispersion, while dielectric measurements and Maxwell-Garnett modeling were used to interpret the observed frequency-dependent permittivity behavior.

2. Materials and Methods

2.1. Materials

HDPE polyethylene granules (SOCAR, "Azerikimya" ProductionUnion "Etilen-Polyethylene" plant, 15803-020), CCl_4 organic solvent (Code 141245, 99.5%, Cas No- [56-23-5], Common Chemistry- P.L.C); tantalum oxide nanoparticle (Ta_2O_5), (Hongwu International Group, Ltd, China, 99.9%, T_50_2), hafnium oxide (HfO_2) nanoparticles, size 10-20nm, Luoyang Tongrun Info Technology Co., Ltd. China, CAS 12055-23-1, 99%).

2.2. Sample Preparation

HDPE/Ta_2O_5 and HDPE/HfO_2 polymer nanocomposites are prepared as follows: polyethylene granules are dissolved at room temperature in $60\ ml$ of carbon tetrachloride (CCl4) at 77°C. Ta_2O_5 nanoparticles are then added to the polymer solution at various volume concentrations and mixed for 8 hours until a homogeneous mixture is obtained. The mixture is transferred to a Petri dish and dried in a vacuum oven for 24 hours. Thin films of the nanocomposites are subsequently obtained by hot pressing the dried samples at the melting temperature of polyethylene and a pressure of $15\ MPa$ for 10 minutes. After hot pressing, the films are cooled in water at room temperature. The film thickness is 95–105 μm.

2.3. Characterization Techniques

High-Voltage CD: The test is designed to evaluate the material's resistance to sudden load application. The CD was generated using "needle-plane" electrodes. The needle diameter is approximately 0.3mm, and the distance between the needles and the film samples is $1\ cm$. The charging voltage $U_c \approx 14.5\ kV$, with a charging time of 10 minutes. The thickness of the samples is $105\mu m$.

Morphology: Scanning electron microscopy (SEM) provided insight into nanoparticle dispersion and aggregate formation [1, 2].

Dielectric Properties: Dielectric constant (ε′) and loss tangent (tan δ) were measu-red using a precision LCR meter across a frequency range of 1 kHz–1 MHz at room temperature [12, 15, 16].

3. Results and Discussion

3.1. High-Voltage Corona Discharge

The corona discharge (CD) method is based on the transfer of charge from the region of electrical discharge in an air (gas) gap to the surface of the composite. During this process, ions either transfer their charge to the composite and return to the air or penetrate the near-surface region of the composite, where they are trapped by ion traps. The CD appears as a glow localized around a point tip within a highly non-uniform electric field. The physics of this phenomenon is well understood-the corona can be considered as a Townsend discharge (a quasi-stationary electrical discharge in a gas) or a negative glow discharge, depending on the field and potential distribution [18]. The dielectric strength of an insulating material is one of the most important electrical properties to determine. HDPE (High Density Polyethylene) has already demonstrated its capabilities in various scientific and engineering applications. Therefore, it is crucial to investigate the breakdown characteristics of HDPE-based nanocomposites to assess their suitability as electrical insulators.

3.2. Scanning Electron Microscopy (SEM)

For HDPE/HfO_2 and HDPE/Ta_2O_5 nanocomposites, SEM and EDS analyses were performed to assess structural and compositional properties. Energy-dispersive X-ray analysis (EDX) using SEM (Figure 1) confirmed the presence of tantalum, hafnium, oxygen, and carbon within the matrix, with weight percentages of Ta (95.49%) and O (4.51%), and Hf (95.55%) and O (4.45%), respectively. The SEM images reveal the distribution of nanoparticles within the HDPE matrix. While elemental mapping indicates a generally uniform dispersion, areas of nanoparticle clustering are observed, particularly in HDPE/5%Ta_2O_5, supporting the TEM findings of more pronounced agglomeration. These differences in dispersion may be attributed to variations in surface chemistry, influencing nanoparticle interaction with the HDPE matrix and the overall effectiveness of the sonication process.

3.3. Dielectric Constant (ε′) of Nanocomposites

The dielectric behavior of HDPE nanocomposites containing HfO_2 and Ta_2O_5 was investigated before and after corona discharge (CD). The dielectric constant (ε) was determined using

$$\varepsilon = \frac{Cd}{\varepsilon_0 A}, \tag{1}$$

where C is the capacitance, d is the sample thickness, ε_0 is the permittivity of free space, and A is the electrode area.

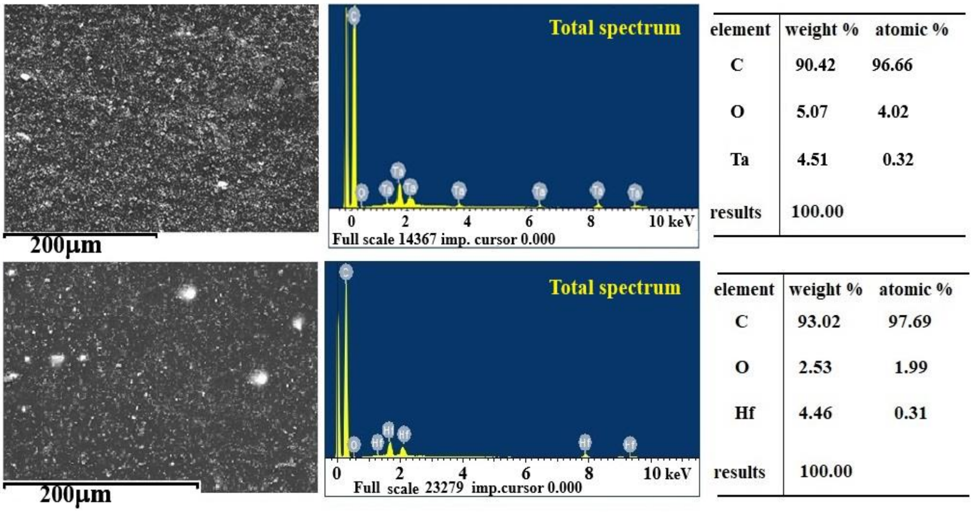

Figure 1. SEM image, corresponding EDX spectra and element mapping of: a) HDPE/HfO_2 and b) HDPE/Ta_2O_5 Nanocomposites

The dielectric constant (εcom) of the nanocomposites was further analyzed using the Maxwell-Garnett model, which predicts how different nanoparticle concentrations influence the dielectric response:

$$\varepsilon_{com} = \varepsilon_m \frac{\varepsilon_f + 2\varepsilon_m + 2f(\varepsilon_f - \varepsilon_m)}{\varepsilon_f + 2\varepsilon_m - f(\varepsilon_f - \varepsilon_m)}. \tag{2}$$

Where εcom is the permittivity of the nanocomposites, ε_m is the permittivity of the matrix phase (HDPE), ε_f is the permittivity of the nanofiller (HfO_2, Ta_2O_5), and f is the volume fraction of the nanofiller. This model helps explain the observed increase in dielectric constant with increasing nanoparticle concentration, as nanoparticles enhance interfacial polarization within the HDPE matrix.

As shown in Figure 2, the dielectric permittivity of HDPE-based nanocomposites remains relatively stable across the frequency range, with higher values at increased filler concentrations. After CD treatment, a slight reduction in permittivity was observed for all samples, likely due to structural rearrangements and altered filler-matrix interfaces. While the Maxwell-Garnett model predicts an increase in ε with filler concentration, the experimental values deviate slightly due to interfacial polarization and charge-trapping effects. The overall decrease in permittivity after CD suggests that the discharge modifies interfacial polarization and charge mobility within the composite. These findings indicate that CD treatment stabilizes the dielectric response and enhances the interfacial compatibility between HDPE and the oxide nanoparticles.

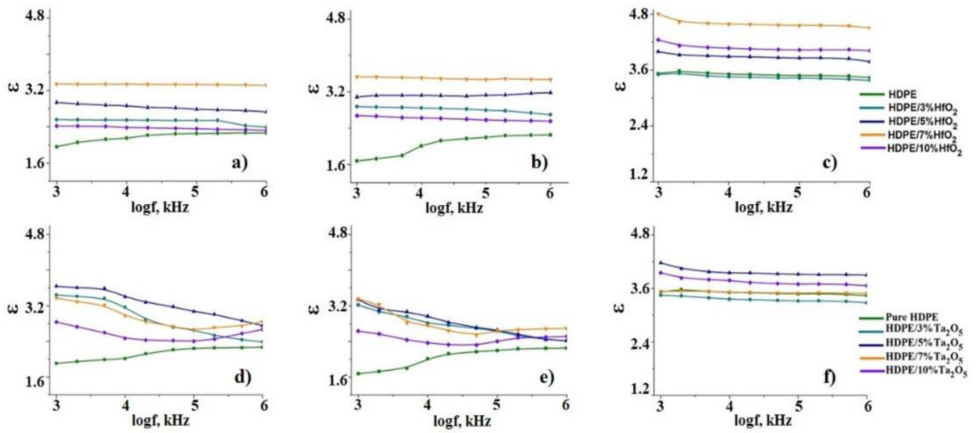

Figure 2. Experimental and theoretical frequency dependence of dielectric permittivity for HDPE/HfO_2 and HDPE/Ta_2O_5 nanocomposites before and after corona discharge (CD).

4. Conclusion

In summary, HDPE nanocomposites containing HfO_2 and Ta_2O_5 nanoparticles were successfully fabricated and evaluated before and after corona discharge (CD) treatment. SEM and EDX analyses confirmed good nanoparticle dispersion with minor agglomeration at higher loadings. The dielectric constant increased with nanoparticle content, consistent with the Maxwell–Garnett model, indicating enhanced interfacial polarization. After CD exposure, a slight decrease in permittivity was observed, attributed to structural rearrangements and modified charge transport at the filler–matrix interface. These results demonstrate that corona discharge effectively alters interfacial polarization and improves dielectric stability. Consequently, HDPE/HfO_2 and HDPE/Ta_2O_5 nanocomposites exhibit strong potential for use in high-voltage insulation systems requiring reliable and durable dielectric performance.

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