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DIELECTRIC PROPERTIES OF MODIFIED THERMOPLASTICS USED FOR 3D PRINTING

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Abstract: This study investigates the temperature–frequency dependencies of the dielectric properties of polymer composite materials (PCMs) based on modified thermoplastics filled with kaolin (AKT-10) particles. Kaolin powders (AKT-10) with effective particle sizes of 2 μm and 20 μm were used as fillers. The structure formation processes of the PCMs were carried out under a magnetic field at elevated temperatures. Temperature–frequency dependencies of the relative dielectric permittivity and the loss tangent were established depending on the filler concentration, particle size distribution, structural organization technologies, and the conditions of magnetic field exposure.

Keywords: filament, dielectric properties, thermoplastic, filler, kaolin

I. INTRODUCTION

Currently, filament for 3D printers is a highly demanded product due to the growing 3D printing market and its widespread application across various industries. It is essential for creating prototypes, small components, artistic objects, as well as for the production of replacement parts [1, 2].

3D printing filament is the material used in 3D printers to create objects. It comes in the form of a thread, similar to sewing machine thread [3].

The popularity of filament is driven by the following factors:

- *Growth of the 3D printing market:*

The consumer 3D printing market is projected to reach $12 billion by 2030.

- *Wide range of applications:*

Filament is used to produce a variety of items, from prototypes and functional parts to decorative objects and even garments.

- *Customization potential:*

3D printing allows the creation of unique, personalized products.

- *Spare parts replacement:*

3D printing is an excellent method for restoring or replacing broken components.

- *Business opportunities:*

3D printing opens the door to creating and selling unique products and offering 3D printing services.

- *Accessibility:*

There are various types of filament suitable for different types of 3D printers and applications [4].
The most commonly used filaments include:

1. *PLA*- An environmentally friendly and easy-to-use material, frequently used for prototypes and models.
2. *ABS*- Durable and heat-resistant, commonly used for manufacturing functional and strong components.

3. *PETG*- Ideal for strong and chemically resistant products, such as containers.

4. *TPU*- A flexible material used for printing elastic parts and prosthetics.

The increasing demand for 3D printing filament is a result of the expanding 3D printing market, a broad spectrum of applications, and the potential for customization and on-demand production. This makes it attractive to a wide range of consumers—from hobbyists to large companies and entrepreneurs [5].

II. RESEARCH METHODS

One of the most important characteristics of 3D printing filament is its electrical conductivity.

To study the electrical conductivity of 3D printer filaments, several methods can be used. The most common among them are **resistance measurement** and **specific (volume) resistivity measurement**. Additionally, methods based on the principles of electromagnetism—such as inductance or capacitance measurements—may be applied, although they are less frequently used in this field [6–8].

Resistance and Specific Resistivity Measurement Methods:

1. Resistance Measurement (Resistivity):

- 1. Prepare a filament sample of the required length and shape (e.g., cylindrical or rectangular).
  2. Use a multimeter or other measuring device to determine the resistance (in ohms).
  3. Ensure reliable contact between the measurement probes and the sample.
  4. Measure the resistance at different temperatures if studying the temperature dependence of conductivity.

2. Specific (Volume) Resistivity Measurement:

- 1. First, measure the sample dimensions (length, width, thickness, or radius).
  2. Then calculate the resistivity using the formula:

$$\rho = \frac{R * A}{L}$$

(where: R - measured resistance (Ω), A - cross-sectional area (m^2), L - length of the sample (m)).

1. (a) Compare the obtained resistivity values with those acquired by other methods to assess measurement accuracy.

Advanced Methods:

1. *Inductance and Capacitance Measurement:*

These techniques may be helpful for analyzing the electromagnetic properties of the filament but typically require specialized equipment.

1. *Hall Effect-Based Methods:*

The Hall effect allows the determination of properties of the conductive medium, such as the type of charge carriers and their concentration.

1. *Spectroscopy-Based Methods:*

Spectroscopy can be used to study the electronic structure of the material and evaluate its electrical conductivity.

1. *Dielectric Property-Based Methods:*

A dielectric tester, such as the **USG-7808** (Figure 1), can be used to automatically determine the dielectric properties of solid dielectrics.Supported standards: ASTM D1816, D877; IEC 156, IEC 60156.The test voltage is selected from a list of standards via the menu or set manually. The results are printed using the built-in microprinter.

Data Processing:

After completing the measurements, it is necessary to process the obtained data for example, by calculating **mean values** and **standard deviations**, as well as by constructing **graphs showing the dependence of resistance on temperature** or other



Figure 1. Dielectric Tester USG-7808.

relevant parameters. It is important to take into account possible measurement errors and their impact on the final results. The obtained data should be compared with known conductivity values of similar materials or with the specifications provided by the filament manufacturer.

Application: Measuring the electrical conductivity of filament can be useful for assessing its compliance with specific requirements or for the development of new material types. Electrical conductivity data can be used to optimize the 3D printing process. For instance, in the development of electrically conductive or magnetically active components manufactured via 3D printing, it is essential to know the conductivity characteristics of the material used.

The selection of a particular method for evaluating conductivity depends on the research objectives and the available equipment. During measurements, it is crucial to follow safety regulations and adhere to proper handling procedures [9].

III. RESULTS AND DISCUSSION

Plastics are inherently good electrical insulators. Moreover, amorphous polymers retain high dielectric properties up to their melting point. Therefore, polymers are often used to manufacture electrical insulators, including those that operate under high-voltage conditions. Some plastics are capable of functioning within a thermal resistance class below 220°C.

The dielectric permittivity of filaments such as PLA + kaolin, ABS + kaolin, PETG + kaolin, and TPU + kaolin was measured using the USG–7808 dielectric tester. The main technical specifications of the tester include:

- test voltage range: 0–100kV,
- electrode spacing: 1 to 3mm,
- tester insulation resistance: not less than 10^6 Ohm [10].

Dielectric Permittivity of PLA + Kaolin Filament

The temperature–frequency dependencies of the relative dielectric permittivity (ε) and the loss tangent ($t_{g\delta}$) of polymer composite materials (PCMs) based on thermoplastics (PLA, ABS, PETG, and TPU) and AKT-10 kaolin filler were investigated. The average particle sizes of the AKT-10 kaolin used were 2μm and 20μm.

The filler content in the samples was varied at 20%, 40%, and 60% by weight. All concentration values indicated in the study refer to mass fractions.

The values of ε and $t_{g\delta}$ were determined using a two-electrode system within a frequency range from 1 to 50kHz, and a temperature range from 295K to 463K. The measurements were conducted using a specially designed shielded and grounded heated test cell of the "sandwich" type, featuring a measuring and potential electrode system with 15mm diameter stainless steel compression electrodes.

Capacitance (C) and loss tangent ($t_{g\delta}$) were measured using a high-precision R5016 device. Based on these measured parameters and the geometric dimensions of the samples, the values of dielectric permittivity (ε) and t_g were calculated using standard methods.

The key results regarding the temperature–frequency and temperature dependence of ε and $t_{g\delta}$ for PLA + kaolin, ABS + kaolin, PETG + kaolin, and TPU + kaolin composite samples are presented in Figures 2–5.

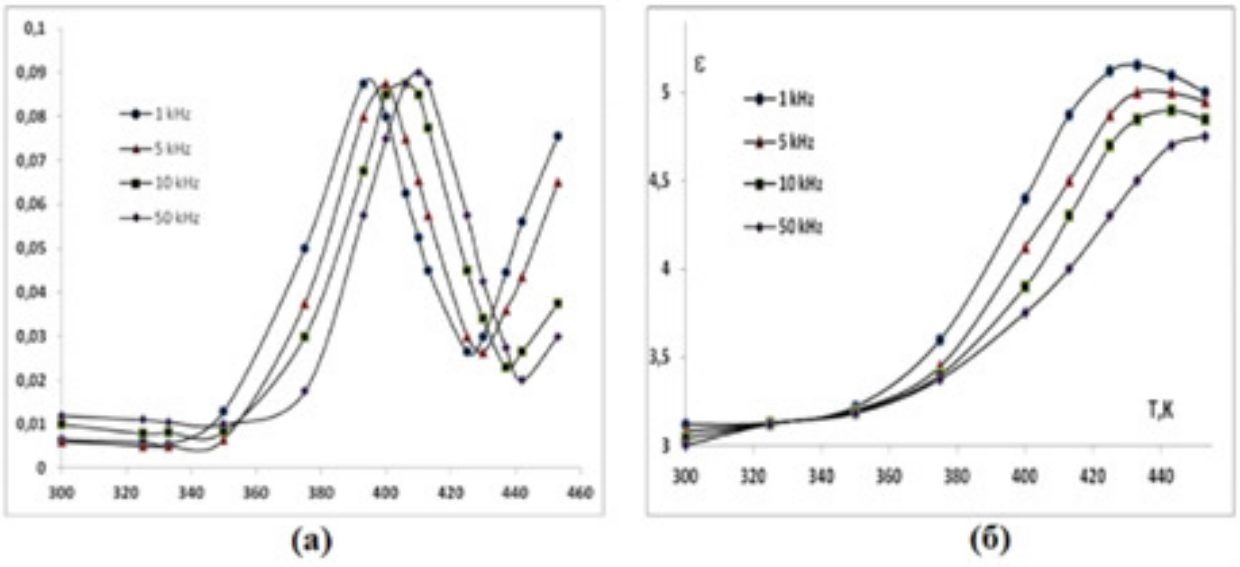

Figure 2. Temperature–frequency dependencies of the dielectric loss tangent ($t_{g\delta}$) and relative dielectric permittivity (ε) of PLA + kaolin (AKT-10)

As shown in Figures 2a and 2b, for PLA + kaolin (AKT-10), the temperature dependence of the dielectric permittivity and dielectric loss tangent ($t_{g\delta}$) exhibits a dipolar–segmental polarization process, manifested by clear peaks at around T=400 K for $t_{g\delta}$ and T = 430 K for ε. As illustrated in the graphs, an increase in temperature leads to a shift of the peaks toward higher frequencies. This behavior is characteristic of the well-known dipolar–segmental polarization, which is associated solely with the oscillations of macromolecular chain segments. The observed peaks correspond to the first relaxation transition.

The subsequent rise in dielectric losses after reaching a minimum is attributed to dipolar–group polarization, caused by the orientation of dipoles belonging to terminal or side groups, as well as to polar molecules of low-molecular-weight additives or impurities that become mobilized at elevated temperatures. At higher temperatures, the viscosity of the polymer matrix decreases, allowing dipoles to reorient freely without resistance.

For the composite samples filled with kaolin (AKT-10) at the given concentration, a noticeable increase in both $t_{g\delta}$ and ε is observed (see Figures 3a and 3b). This effect can be explained by the growth of crystalline regions in the polymer matrix, induced by the crystalline structure of the kaolin particles, which enhances the measured dielectric parameters.

However, the temperature dependence of $t_{g\delta}$ varies depending on the particle size of kaolin (AKT-10):

1. For finer particles (d = 2μm), $t_{g\delta}$ behaves similarly to that of the pure polymer, displaying two relaxation peaks (Figure 3a). The first, weakly expressed peak appears around T = 380K, while the second, more prominent peak emerges at T > 400K. These peaks shift to higher temperatures and decrease in amplitude as the frequency increases. This is because, at higher frequencies, dipoles and ions do not have enough time to follow the electric field oscillations, leading to reduced polarization and a sharp drop in $t_{g\delta}$.

2. For coarser particles (d = 20μm), a more pronounced growth in $t_{g\delta}$ is observed with temperature. A broad, weak maximum occurs in the range of T = 373–413K, followed by a significant increase in $t_{g\delta}$ at higher temperatures (Figure 3b).

The observed differences in the behavior of $t_{g\delta}$ can be attributed, in the first case, to the high defectiveness of the initial structure and surface of the barium hexaferrite particles with a size of d = 2μm. In the second case, the particles with d = 20μm likely have a more densely packed crystalline structure, and as the temperature increases, the intensity of charge displacement or migration also increases. In this scenario, dielectric losses are primarily associated with conductivity, which leads to the observed pronounced growth of $t_{g\delta}$.

These interpretations of the dielectric permittivity and loss behavior are further supported by the subsequent measurements of dielectric parameters at varying concentrations of kaolin (AKT-10), presented in Figures 4 and 5.

It is well known that kaolin exhibits a relatively high dielectric permittivity, which depends on both frequency and compositional structure. As the frequency increases, the dielectric permittivity of kaolin (AKT-10) decreases, and a magnetostriction effect may be observed within the material.

As shown in Figure 4, at equal concentrations of kaolin (AKT-10), the highest values of ε are observed in PCMs filled with 2μm particles. This can be explained by the higher defectiveness of the crystalline structure in these particles. The more ionic

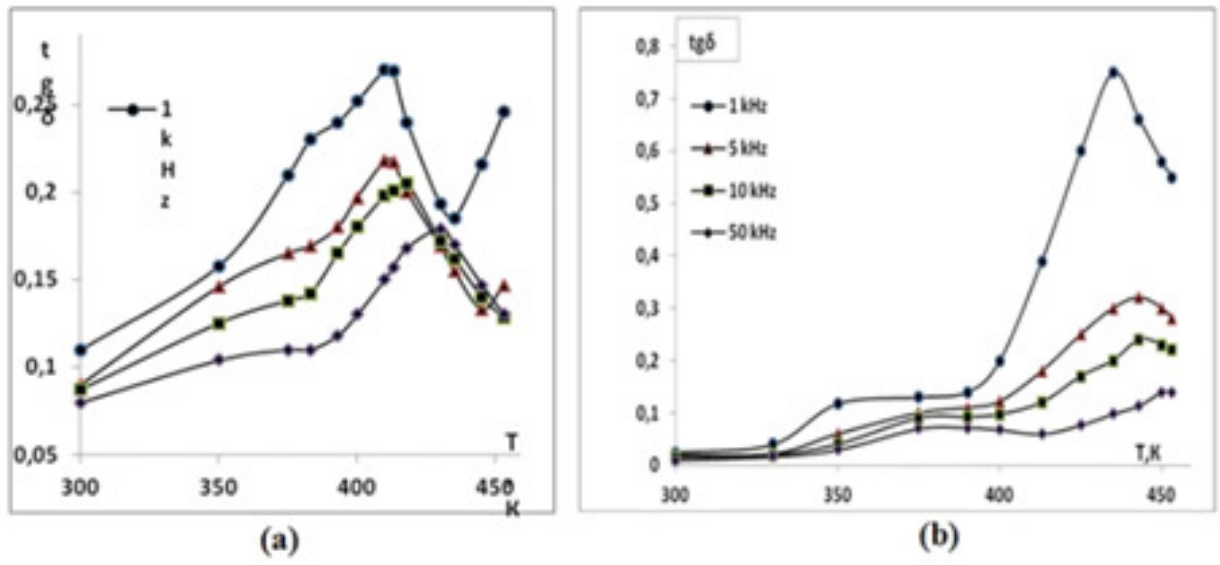

Figure 3. Temperature–frequency dependencies of the dielectric loss tangent ($t_{g\delta}$) and relative dielectric permittivity (ε) for ABS + kaolin (AKT-10)

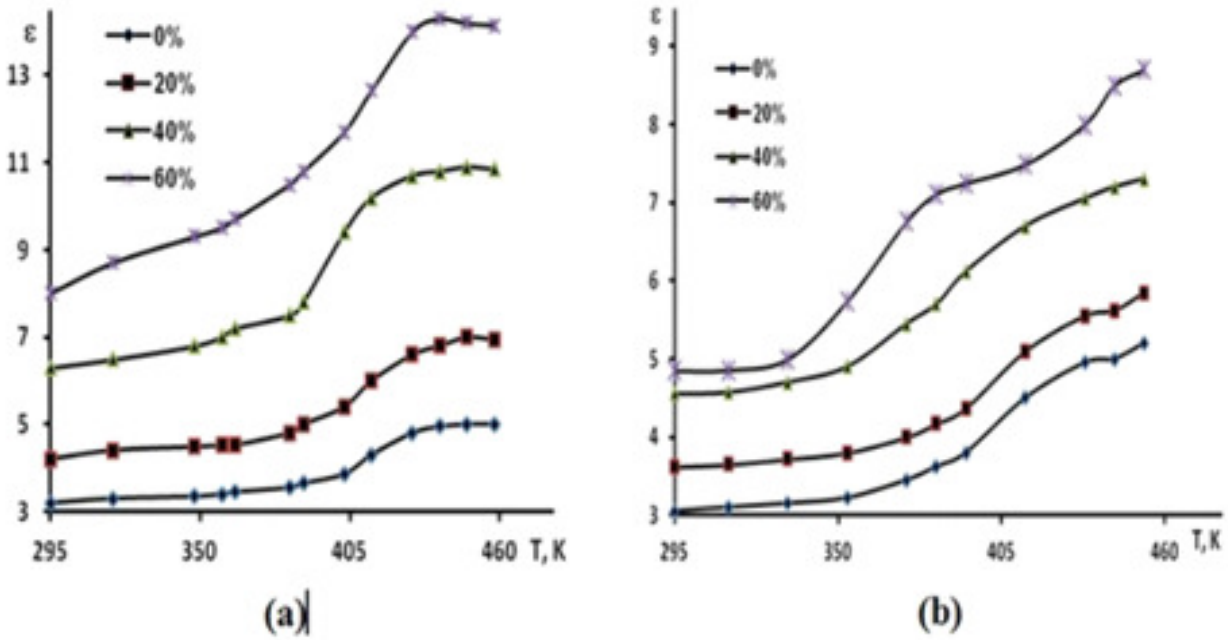

Figure 4. Temperature–frequency dependencies of the dielectric loss tangent ($t_{g\delta}$) and relative dielectric permittivity (ε) for PETG + kaolin (AKT-10)

vacancies and weakly bound electrons present in the kaolin structure, the more dipoles are generated, resulting in a higher dielectric permittivity.

The weak peaks observed in the dielectric spectra are related to disorientation processes of electronic, ionic, ion-relaxational, and migrational polarization, which occur around T = 438K. The contribution of each mechanism to the overall polarization ultimately depends on the lattice energy of kaolin's crystalline structure and the energy state of its specific surface area.

The characteristic behavior of ε in Figure 4a for T $>$ 405K and kaolin particles with d = 2μm is explained by the increased intensity of free charge movement. At elevated temperatures, viscosity decreases, allowing dipoles to shift freely without friction.

The high defectiveness of the structure and surface contact of kaolin (AKT-10) particles with a size of d = 2μm is also evident in the behavior of $t_{g\delta}$ (see Figure 5a).

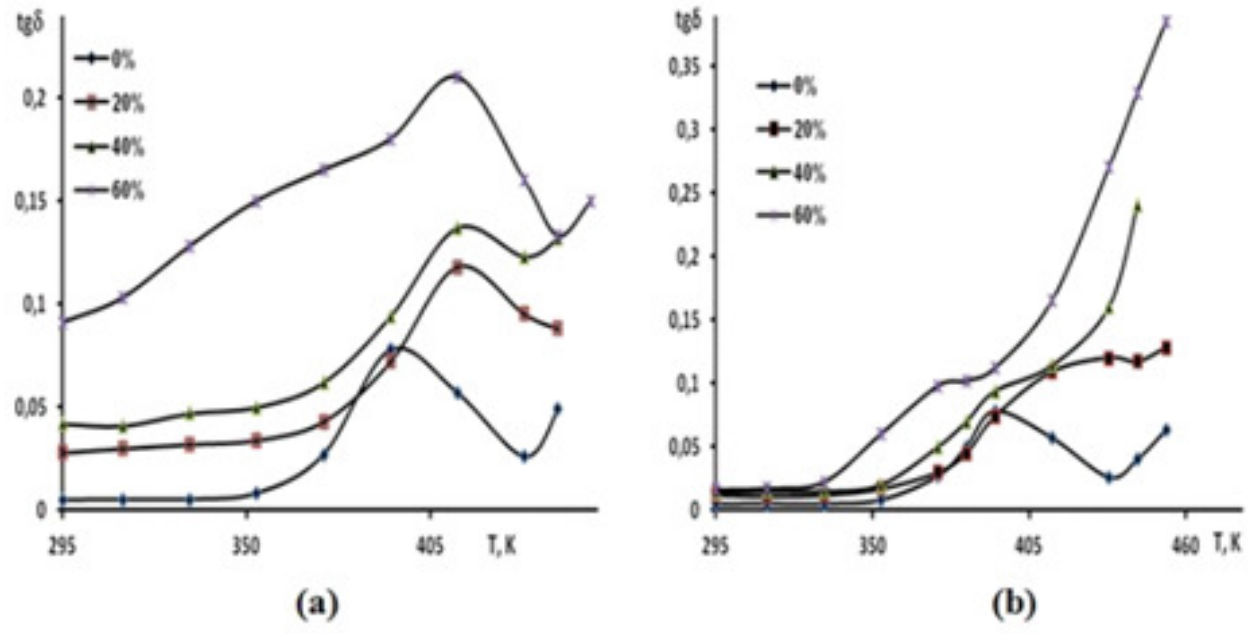

Figure 5. Temperature–frequency dependencies of the dielectric loss tangent ($t_{g\delta}$) and relative dielectric permittivity (ε) for TPU + kaolin (AKT-10)

As shown in the temperature dependence of the dielectric loss, the value of $t_{g\delta}$ in the PCM increases with the rising concentration of filler particles, and the relaxation transition peaks shift toward higher temperatures. This effect may be attributed to the formation of interfacial layers at the contact boundary between the polymer matrix and the surface of the filler particles. As a result, the number of free macromolecular segments participating in polarization processes in the pure polymer is reduced.

The minima observed in Figure 5a at T = 433–453K, followed by a further increase in $t_{g\delta}$, can be associated with the growth of migrational and other types of polarization due to the accumulation and eventual saturation of charge states at these temperatures. This may be a consequence of changes in the crystalline structure and the initial defectiveness of the kaolin (AKT-10) particles' contact surface with d = 2μm.

The temperature and frequency dependencies of the dielectric permittivity and dielectric losses for PCM samples filled with kaolin (AKT-10) particles of average size d = 20μm show behavior typical of dielectrics with a dominant hopping conduction mechanism (Figures 4b and 5b). This is indicated by a notable decrease in relative dielectric permittivity and a sharp increase in dielectric loss with increasing temperature.

It is known that as frequency increases, the dielectric permittivity of kaolin (AKT-10) decreases, while with increasing temperature, its conductivity improves.

Thus, kaolin (AKT-10) particles with d = 20μm are characterized by higher crystalline packing density and lower surface defectiveness compared to particles with d = 2μm. Therefore, the weak increase in ε may be attributed to the manifestation of migrational polarization (Figure 3b), as well as the presence of a glassy interfacial layer on the surface of these particles.

IV. CONCLUSION

Using electromagnetic technology, polymer composite material (PCM) samples were fabricated, filled with varying concentrations of kaolin (AKT-10) particles with dispersions of 2μm and 20μm.

Uniform distribution of kaolin (AKT-10) particles within the polymer matrix was achieved by rotating the filled compound during thermal curing in a constant magnetic field.

The dielectric properties of the synthesized PCM samples were investigated in the low-frequency range (1kHz – 50kHz) and within the temperature range of 300–463K.

Temperature–frequency dependencies of the relative dielectric permittivity (ε) and dielectric loss tangent ($t_{g\delta}$) were obtained, followed by a detailed discussion, explanation, and scientific interpretation of the observed changes in dielectric behavior.

The influence of highly dispersed kaolin (AKT-10) particles with average sizes of 2μm and 20μm on the values of ε and $t_{g\delta}$ in PCMs was established. This behavior is attributed to the crystalline structure condition of the kaolin filler.

It was demonstrated that mechanical milling may introduce defects into the original kaolin (AKT-10) structure, affecting its

dielectric properties. This effect is more pronounced for particles with d = 2μm and at filler concentrations up to 60 parts by mass in the polymer matrix.

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