

폴리스타이렌@바륨타이타네이트 입자 배열과 용착을 통한 임베디드 커패시터용 고유전 박막 형성

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Formation of High-Dielectric Layers through Annealing of Polystyrene@Barium Titanate Arrays for Embedded Capacitor

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초록: 고유전 박막은 전자회로의 전하저장 소자의 성능을 결정하는 중요한 소재로서, 임베디드 커패시터에서는 고분자와 고유전 필러의 복합막을 통해 구현된다. 이에 대한 효율적인 제조 방법으로서, 폴리스타이렌@바륨타이타네이트(PS@BT) 입자를 제조하고 이를 배열한 층을 드롭 캐스팅으로 형성한 후 열적으로 용착하여 고유전 복합막을 형성하였다. 복합막의 단면 관찰에 의하면 BT 입자는 PS 매트릭스와의 층분리나 응집 없이 균일하게 분포된 막을 형성할 수 있음을 확인하였다. 유전특성 조사에서는 BT 함량 55.6 wt%(PS@BT-1)와 65.7 wt%(PS@BT-2)인 박막에 대해 각각 30.9과 36.2의 유전상수값을 얻을 수 있었고, 누설전류밀도는 각각 5.98과 496 nA/cm²를 나타내었다. 두 BT 함량 사이에서 누설전류의 퍼콜레이션 포인트(percolation point)가 형성됨을 확인할 수 있었다. 126 V/μm의 높은 절연과과전압을 나타내는 PS@BT-1 박막에 대해서는 2.17 J/cm³의 에너지 저장밀도를 확보하였다.

Abstract: High-dielectric thin films are crucial materials that determine the performance of charge storage devices in electronic circuits, particularly in embedded capacitors where they are realized through composite films of polymers and high-dielectric ceramic fillers. As an efficient fabrication method, polystyrene@barium titanate (PS@BT) particles were synthesized, arranged into layers *via* drop casting, and then thermally fused to form high-dielectric composite films. Cross-sectional observations of the composite films revealed that BT particles were uniformly distributed within the PS matrix without any layer separation or agglomeration. Dielectric property investigations showed that thin films with BT contents of 55.6 wt% (PS@BT-1) and 65.7 wt% (PS@BT-2) exhibited dielectric constants of 30.9 and 36.2, respectively, with leakage current densities of 5.98 and 496 nA/cm². It was confirmed that a percolation point for leakage current existed between these BT content levels. For the PS@BT-1 film, which demonstrated a high breakdown voltage of 126 V/μm, an energy storage density of 2.17 J/cm³ was achieved.

Keywords: polymer, barium titanate, high-dielectric, capacitor, nanocomposites.

Introduction

Capacitors are fundamental components in electronics, serving as reservoirs that store and release electrical energy, which has gained significant attention in recent years and is extensively utilized in several industries, including automotive, electrical devices, and defense systems.¹⁻⁴

Their properties include capacitance, which is the ability to store an electric charge, and working voltage, which defines the maximum voltage they can handle and their efficiency. With the advancement of the electronics industry, there has been a significant trend towards miniaturization, lightweight, and enhanced functionality of electronic devices. As a result, proactive efforts are being undertaken to design capacitors that can efficiently store energy within these devices.^{5,6}

High dielectric constant materials play a crucial role in the development of capacitor devices due to their ability to store

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electrical energy efficiently. The dielectric constant, also known as the relative permittivity of a material, is a measure of its ability to concentrate electric flux and is directly proportional to the capacitance of a capacitor.

Ceramic materials are widely used to manufacture capacitors due to their dielectric properties, which allow them to store a significant amount of electrical energy relative to their size.⁷⁻⁹

The high dielectric constant of ceramic materials enables the production of capacitors with high capacitance values while maintaining compact dimensions. Moreover, ceramic capacitors exhibit stability over a wide range of temperatures and frequencies, making them suitable for diverse applications. Barium titanate (BT), a ferroelectric ceramic, is known for its high dielectric permittivity, making it highly suitable for use in multilayer ceramic capacitors.¹⁰⁻¹² BT capacitors find applications in various electronic circuits, offering advantages like compact size and stable performance. Despite its benefits, BT does have limitations, including nonlinear temperature coefficients and potential brittleness, which can lead to mechanical failures.^{13,14}

Integrating BT with polymers to create composites has emerged as a solution to mitigate some of these issues. By combining BT with a polymer matrix, such as poly(methyl methacrylate) (PMMA),¹⁵ epoxy,¹⁶ polyimide,¹⁷ and poly(vinylidene fluoride) (PVDF),¹⁸ the resulting composite material can exhibit improved flexibility, processability, and high breakdown resistance compared to pure ceramic capacitors. However, these composites have the disadvantage that when ceramic is added at a high percentage, the surface roughness of the film increases due to aggregation and inhomogeneous phase formation, and the process strength of the polymer is weakened, resulting in a decrease in properties such as dielectric properties thermal stress reliability.^{19,20} In general, the dielectric properties of polymer/ceramic composites can be effectively improved by composited morphology, and the dispersion and connectivity of the components in the polymer matrix are important factors that can improve the dielectric properties.^{12,21}

In this study, we propose a fabrication method for the dielectric composite films to minimize the aggregation and separation of high-dielectric ceramic particles, which are the primary causes of dielectric property deterioration in polymer/ceramic particle composites. To achieve this, we synthesized PS@BT by attaching BT to sub-micron PS particles and then formed a layer of particle array through a simple drop-casting method. By applying heat treatment near the glass transition temperature, we created a uniform film to form the dielectric composite layer. This approach addresses the common issue of polymer-filler sep-

aration during the formation of high-dielectric composite layers and offers a novel fabrication method that contrasts with traditional methods, which often involve the use of special compatibilizers to manage the polymer-filler interface and affect the dielectric properties. The uniform and stable high-dielectric filler/polymer composite layer is expected to play a crucial role in providing a high-energy density thin film for embedded capacitors.

Experimental

Materials. Styrene, potassium persulfate (KPS) (99%), and barium titanate (99%) were purchased from Sigma-Aldrich. Ethanol and toluene were provided by Alfa Aesar.

Synthesis of Polystyrene@BT Nanoparticles. Polystyrene (PS) colloids were synthesized *via* emulsion polymerization using styrene as a monomer and potassium persulfate (KPS) as an initiator. At first, a 500 mL round bottom flask was charged with 150 mL of D.I water and covered the mouth with septa. Nitrogen gas was bubbled during the reaction to remove moisture. 10.0 g (0.09 mol) of styrene monomer was dropwise and stirred slowly. Then, the temperature was increased to 85 °C, and 0.014 g of KPS in 10 mL D.I water was added into the flask. The mixture was stirred at 85 °C for 24 h to obtain a white polystyrene emulsion solution. D.I water was removed from the obtained emulsion to collect PS as a white powder.

After PS was prepared, an appropriate amount of barium titanate (BT) and the obtained PS powders with weight ratios of PS:BT=1:1, 1:2, 1:3 were mixed together to make a 10 wt% solution in ethanol. Then, the BT nanoparticles were physically adsorbed onto the surface of the polystyrene by sonication treatment for 1 hour. With majority amount of BT particles adhered on the PS surface and without the formation of by-product, the obtained PS@BT solution can be directly used in the next drop-casting process. The films of the dielectric composites were formed by dropping 10 wt% PS@BT solution in ethanol on a clean glass plate and drying before heat treatment. As-prepared films were then obtained after thermal treatment at 100 °C for 30 min to ensure good compactness in the final samples. After the first layer was uniformly dried, the second PS (10 wt% in toluene) layer was cast onto the first layer, followed by 60 min drying at 100 °C. The film thickness was controlled by an applicator, and the final thickness was about 10 μm.

Characterization of Composites. The morphology and cross-sectional structure observation of the samples was carried out

using scanning electron microscopy (SEM, JEOL JSM-6701F, Japan). The composition of PS@BT was characterized by thermogravimetric analysis (TGA, Q5000 TA Instruments, USA) at a heating rate of 10 °C per minute from 25 to 800 °C under oxygen.

Gold electrodes were deposited on both sides of the films at a rate of 10 Å/s using a thermal evaporator (JVR-72S, Jvac, Korea) for the measurement of dielectric properties. The electrical properties of the dielectric materials were measured at the frequency range from 100 Hz to 1 MHz using a precision impedance analyzer (4294A, Agilent Technologies, USA). The leakage current was measured using an Agilent E5272A. The breakdown strength was measured by a high-voltage source meter (Keithley 2410, Keithley Instruments, USA).

Results and Discussion

PS sub-micron particles were prepared *via* the emulsion polymerization method. Potassium persulfate was used as a radical polymerization initiator to form spherical PS in a colloidal state. The spherical morphology of the synthesized PS nanoparticles was confirmed by SEM, as shown in Figure 2(a). Uniform spherical PS particles were formed without aggregation, having an average size of about 300 nm.

To fabricate the PS@BT nanocomposites, PS particles were employed as a matrix to prevent the aggregation of BT particles, which are noted for their high dielectric properties. The detailed procedure for fabricating the PS@BT composites

and the corresponding films is comprehensively illustrated in Figure 1.

Physical adsorption rather than chemical reactions primarily drove the adsorption mechanism of ceramic nanoparticles onto the PS matrix. During the polymerization of polystyrene monomer, potassium persulfate decomposes in water, yielding sulfate radicals that initiate the polymerization process and attach to the surface of the growing polystyrene particles. This method results in PS particles with distinct surface properties attributed the presence of sulfate groups. As a result, the surface of the polystyrene particles will carry negative charges due to the presence of these sulfate groups. In addition, BT can possess surface charges depending on its surface treatment and the pH of the surrounding medium. The true isoelectric point (IEP) value of BT is in the neutral pH range (6-7) and is increased to approximately 9 with 10 wt% BT solution.²² The surrounding medium—ethanol with a pH around 7.33—further contributes to the positively charged surface of the BT particles. Consequently, these positively charged BT particles exhibit electrostatic attraction toward the negatively charged polystyrene particles.²³ Specifically, BT particles were physically adsorbed onto the surface of polystyrene particles through sonication for a duration of less than one hour in an ethanol solution. It is crucial to note that extending the sonication treatment beyond one hour induced the aggregation of BT particles, which distanced them from the polystyrene surface. This phenomenon highlights the delicate balance required in the sonication process to maintain optimal particle dispersion.

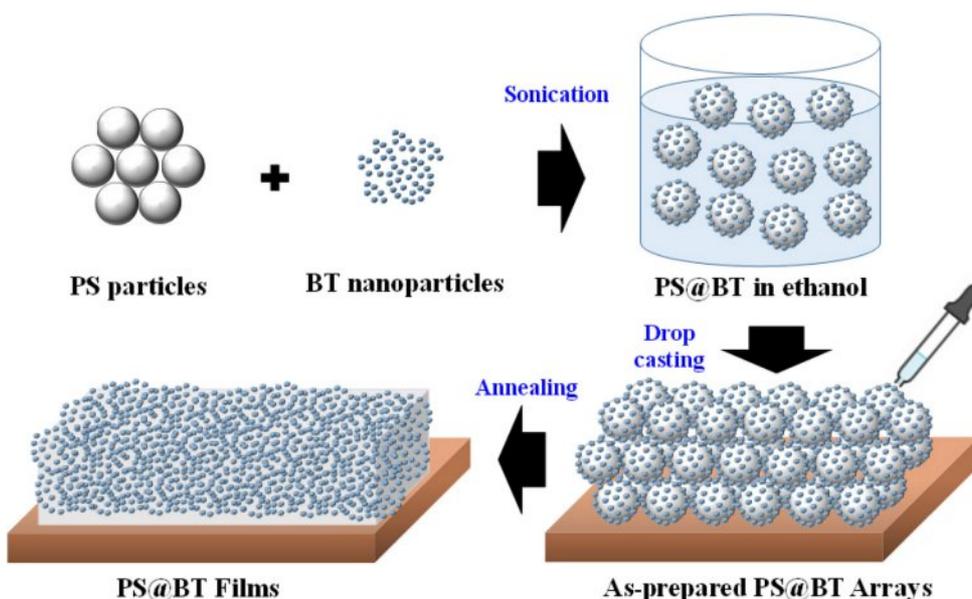


Figure 1. Schematic illustration of PS@BT composites preparation process.

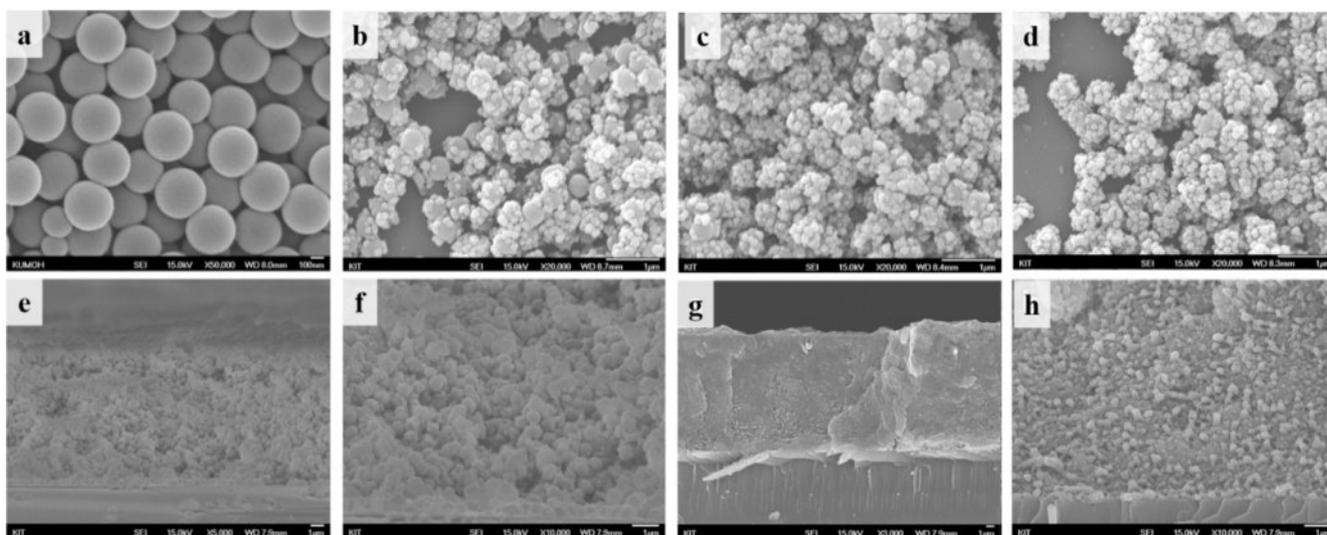


Figure 2. FE-SEM images of (a) synthesized PS particles; (b) PS@BT-1; (c) PS@BT-2; (d) PS@BT-3 composites (with PS:BT weight ratio of 1:1, 1:2, 1:3, respectively). Cross-section FE-SEM images of PS@BT-1 film; (e,f) before heat treatment; (g,h) after heating at 100 °C for 30 min.

To comprehensively evaluate the degree of surface adsorption of BT nanoparticles onto the polystyrene, the morphological evolution of PS@BT particles was monitored. Figure 2(b)–(d) presents the FE-SEM images of the sample as the weight ratio of BT particles in PS increases, thereby augmenting the proportion of high-conductivity ceramics within the polymer matrix. It was observed that BT nanoparticles dispersed uniformly on the polystyrene surface without forming clusters. As the BT content increased, the quantity of BT particles adhering to the PS surface correspondingly rose. The optimal adsorption of BT onto the PS surface without significant BT particle agglomeration was achievable at the PS:BT weight ratio of 1:2. Above this threshold, the limited surface area of the polystyrene constrained further BT adsorption, resulting in the aggregation of surplus BT particles (Figure 2(d)).

The mass of BT particles attached to the polystyrene surface was verified through thermogravimetric analysis (TGA) curves, as illustrated in Figure 3. During the heating process of the PS@BT nanocomposites, polystyrene, which possesses a lower decomposition temperature, decomposes initially, whereas the BT particles, with their higher thermal stability, remain intact. This differential decomposition behavior allows for precisely quantifying BT content within the composites. For the PS@BT-1, PS@BT-2, and PS@BT-3 samples, complete decomposition of the polystyrene matrix was observed at temperatures exceeding approximately 400 °C. The final residues, which correspond to the remaining BT particles,²⁴ were measured at 55.6, 65.7, and 78.3 wt%, respectively.

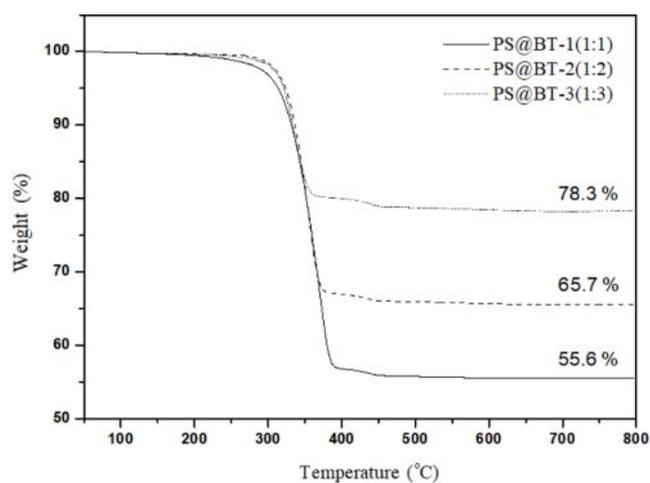


Figure 3. TGA of PS@BT nanoparticles.

These residue percentages directly correlate with the initial BT particle content in the nanocomposites, confirming the successful incorporation and retention of BT particles within the polystyrene matrix at the specified weight ratios. Despite its high dielectric constant, BT is limited in its application as an energy storage material due to its low processability and lack of flexibility. A film of the dielectric material was formed using the solution-processable polymer polystyrene as a matrix to address these shortcomings. The uniform dispersion of ceramic particles within the polystyrene matrix is anticipated to improve the dielectric constant of the resulting films and enhance the overall functionality of the composite.

Figure 2(e)–(h) shows FE-SEM images of fractured cross-

sections of the as-prepared films before and after heat treatment. Before heat treatment, the PS@BT-1 film exhibited a homogeneous dispersion of BT nanoparticles within the PS matrix, with indistinct boundaries between BT and PS. Uniform polystyrene particles that are packed in a hexagonal pattern can decrease the gap between layers when creating the particle array. This, in turn, enhances the even distribution of BT particles in the composite material. To achieve a fully fused layer of polystyrene particles, a heat treatment was conducted at a temperature of 100 °C for 30 min, taking into account the glass transition temperature (T_g) of PS, which is around 100 °C. The ceramic particles are uniformly distributed without any concentrated clustering, and the polystyrene sphere structure, which serves as the matrix, undergoes deformation during heat treatment of the PS@BT nanocomposite. Thus, the potential formation of pores between the particles, which causes a reduction in dielectric properties, was prevented.²⁵ The initially rough surface of the PS@BT array (Figure 2(e)) was converted into a smooth surface in the composite film, as shown in Figure 2(g). A fully integrated PS@BT was achieved following the PS coating process. The devices were fabricated in parallel-type embedded capacitors to investigate the dielectric properties of PS@BT nanocomposites. The conducting electrodes on both sides were made of gold and had a thickness of 70 nm (Figure 4).

The PS@BT nanocomposites used for device fabrication were PS@BT-1 and PS@BT-2, containing 55.6 wt% and 65.7 wt% BT, respectively, which adsorb well to the surface of poly-

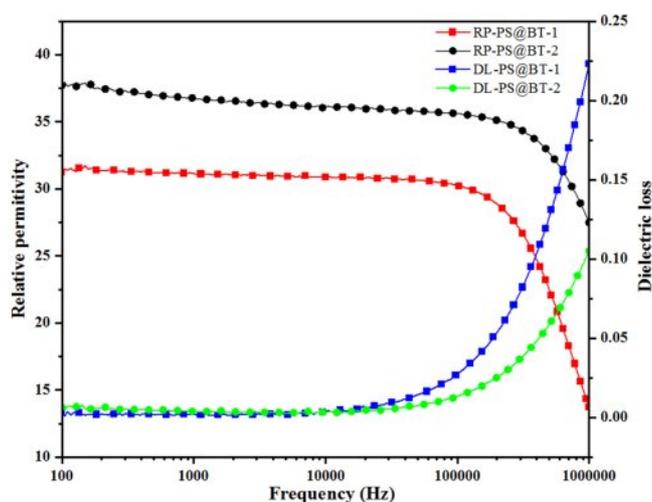


Figure 5. The dielectric constant and dielectric loss of PS@BT nanoparticle films as a function of BT content.

styrene without aggregation of BT particles. The relative permittivity was measured in the frequency range from 100 Hz to 1 MHz. Figure 5 shows the relative permittivity of the two PS@BT nanocomposite films as a function of BT content, which shows an improvement in permittivity compared to the low permittivity of pure polystyrene ($\epsilon_r = 2.5-2.6$). PS@BT-1 and PS@BT-2 films containing 55.6 wt% and 65.7 wt% BT exhibit dielectric constants of 30.9 and 36.2 at 10 kHz, respectively. An increase in relative permittivity was observed for nanocomposite films with a higher content of BT in PS@BT-2 since

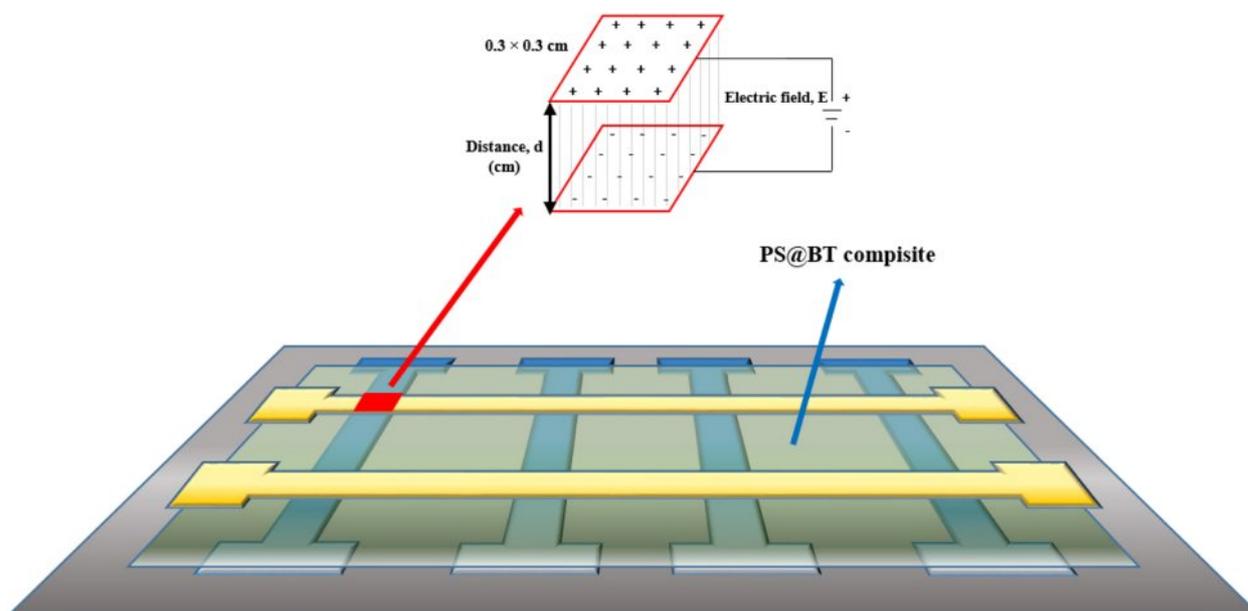


Figure 4. Embedded capacitor of parallel type fabricated with PS@BT nanocomposite film as dielectric.

the relative permittivity is dependent on the dielectric material content of nanocomposites.

However, despite the high content of BT with a high dielectric constant, the improvement in the dielectric constant is also influenced by the dispersion of the ceramic particles, resulting in only a slight increase. In polymer/ceramic dielectric materials, the dispersion of inorganic particles is an essential factor in exhibiting a high dielectric constant. Generally, dispersing reagents increase the dispersibility of inorganic particles in polymers to enhance their dielectric properties. In the case of PS@BT nanoparticles in the prepared films, the inorganic particles were physically adsorbed on the polymer surface without using dispersion-enhancing agents to overcome the manufacturing shortcomings.

Figure 6 illustrates the leakage current density measurements for PS@BT nanoparticle films with increasing electric field. At 0.02 MV/cm, PS@BT-1 and PS@BT-2 exhibited 5.98 and 495.5 nA/cm² values, respectively. The relatively higher BT content increased the leakage current value for PS@BT-2 nanoparticle film. It can be assumed that the surface interaction between BT nanoparticles influenced the development of the current pathway. From this, it is inferred that the percolation point is formed within this range of BT content due to the high BT concentration. In general, ceramics with high dielectric constant values exhibit high dielectric loss characteristics at high electric fields. Obtained dielectric loss values at 10 kHz were 0.02 and 0.02 for PS@BT-1 and PS@BT-2, respectively.

The dielectric breakdown (E_b) of PS@BT-1, a key parameter to calculate the energy density of composites, showed a value

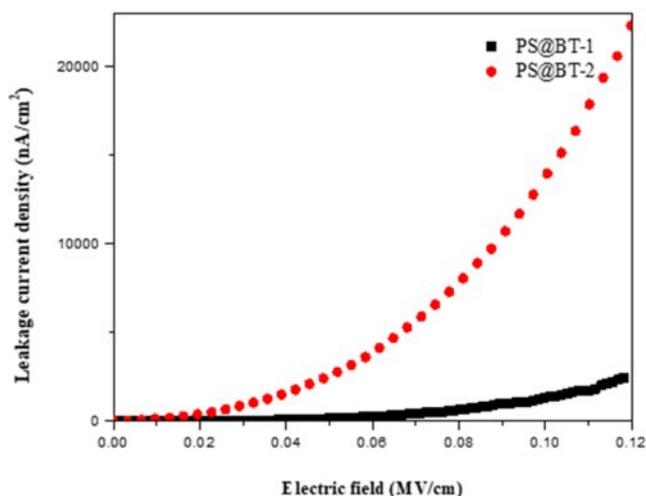


Figure 6. Leakage current density of PS@BT film as a function of BT content.

Table 1. Dielectric Properties of Polystyrene@BT Nanoparticle Films

	PS@BT-1 (TGA 55.6 wt%)	PS@BT-2 (TGA 65.7 wt%)
Film thickness (μm)	7.62	3.09
Relative permittivity ϵ_r (at 10 kHz)	30.9	36.2
Leakage current density (nA/cm ²) ^a	5.98	495.5
Dielectric loss (at 10 kHz)	0.02	0.02
Breakdown strength (V/ μm)	126	-
Energy density (J/cm ³) ^b	2.17	-

^aMeasured at 0.02 MV/cm for 9 mm² area). ^bEnergy density is calculated using $U = 0.5\epsilon_0\epsilon_r E_b^2$ (E_b , breakdown strength (MV/cm)).

of 126 V/ μm , but for PS@BT-2, which has a significantly higher leakage current density value, the dielectric breakdown value could not be obtained due to the high current value. When the energy density was calculated using the equation $U = 0.5\epsilon_0\epsilon_r E_b^2$,²⁴ where ϵ_0 is the vacuum permittivity (8.8542×10^{-12} F/m) and ϵ_r is the relative dielectric constant, it was found that the energy density of PS@BT-1 had a value of 2.17 J/cm³.

Conclusions

In this study, a strategy to produce polymer/ceramic composites with improved dielectric constants for energy storage devices was proposed. Uniform 300 nm-sized polystyrene PS particles were prepared and used to synthesize PS@BT particles *via* attachment of BT particles on PS particles. The layers of PS@BT arrays were fabricated using the drop-casting method and heat treatment to form a fused PS matrix containing dispersed BT particles. SEM images illustrated that BT particles were uniformly distributed in PS matrix without aggregation. The obtained PS@BT composite layers show improved dielectric properties. The relative permittivity of PS@BT-1 and PS@BT-2 films containing 55.6 wt% and 65.7 wt% of BT particles exhibited values of 30.9 and 36.2, respectively. The PS@BT-1 film exhibited low leakage current, high dielectric constant, and high breakdown voltage, resulting in an energy storage density of 2.17 J/cm³. This demonstrates that the fabrication method through forming an array of PS@BT particles and thermally fusing them can provide polymer composite layers with high energy storage density for embedded capacitors.

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Conflict of Interest: The authors state that there is not at all conflict of interest.

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