

Action of colloidal silica films on different nano-composites



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ABSTRACT

Nano-composite films have been the subject of extensive work to develop the energy-storage efficiency of electrostatic capacitors. Factors such as polymer purity, nano-particles size, and film morphology drastically affect the electrostatic efficiency of the dielectric material that form an insulating film between conductive electrodes of a capacitor. This in turn affects the energy storage performance of the capacitor. In the present work, we have studied the dielectric properties of 4 high pure amorphous polymer films: polymethylmethacrylate (PMMA), polystyrene, polyimide and poly-4-vinylpyridine. Comparison between the dielectric properties of these polymers has revealed that the higher break down performance is a character of polyimide PI and PMMA. Also, our experimental data shows that adding colloidal silica to PMMA and PI leads to a net decrease in the dielectric properties compared to the pure polymer.

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Introduction

High operating-voltages are an essential character to assess and to improve the efficiency of the energy storage of materials. The dielectric properties of insulating materials separating the electrodes of a capacitor control the maximum electrostatic power of a capacitor. As an example, polypropylene (biaxial-oriented) has high electrical break down that reached about 6×10^8 V/m; however, it has limited energy storage that reaches about 2 J/cm^3 . A solution to improve the energy storage efficiency is adding nano composite polymers [1–4]. Morphology of the fillers at nanoscale reduces heterogeneities in Nano composites and thus improves the dielectric properties and avoids failure. In addition, to increase these properties, several authors have suggested adding of metal-oxide nanoparticles; of TiO_2 and BaTiO_3 [5–10]. In fact, the dielectric properties are very sensitive to the addition of these nanoparticles because the energy density has quadratic dependence on electric field intensity. Monotonic reduction of the electrical break down appears by increasing the metal-oxide nanoparticles [11–17]. This can be analyzed considering randomly dispersed nanoparticles which form percolation networks which can prematurely break down the material, in particular at high loading rates of TiO_2 and BaTiO_3 . At such rates, it is not a facile task to achieve homogeneous dispersions but it is more likely the formation of

agglomeration that can behave as defect sites leading to more reduction of dielectric properties. Published studies [18–21] have revealed that localized electric fields are created within the matrix with field exclusion. For example addition of (5–10% v/v) BaTiO_3 -nanoparticles generates local electrical fields that negatively affect dielectric properties [20,21]. So, material morphology and individual field-contributions are substantial factors that affect dielectric strength and electrical breakdown. Here, the nature of filler materials can decrease field exclusion and gives deeper insight to the effect of morphology on electrical break down values. As an example to this effect, addition of 5% w/w silica nano particles raises the electrical break down from 2.69×10^8 V/m up to 3.14×10^8 V/m [22,23] and addition of 1% silica to polypropylene increases electrical break down from 5.11×10^8 V/m up to 7.78×10^8 V/m [24]. In addition, the size of filler strongly affects the break down values [25–27] and increasing of these values can occur with more homogenization of the local electric field distribution through the material [28,29]. However, other authors have proposed that the rise of break down values is essentially due to the presence of some internal barriers through the material which can be reduced by good control of film-morphology. If one forces the electrical field to pass through more curved paths a prolonged break down will be produced through tightly structured films [30]. From the experimental point of view, alternating nano-layers of high dielectric polymer: poly (vinylidene fluoride) (PVDF) and polycarbonate have increased dielectric strength which is created by the barriers present in the polymer. These barriers, in turn impede the transfer of electric field [31–34]. Moreover, polyvinyl butyral (PVB) which is added to organically modified montmorillonite (oMMT) in nano

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laminar structures have increased breakdown strength 1×10^8 V/m (pure film) up to 1.3×10^8 V/m [35,36] when adding 10% v/v oMMT. Also, if the filler substance adopts long range alignment, breakdown through oMMT-polyethylene Nano composites will increase. For these films, breakdown strength increases 2.9×10^8 V/m (pure film) up to 3.7×10^8 V/m [36] when adding 6% w/w oMMT. Thus, in general, filler composition, morphology and size are essential factors to get a preferment dielectric material and to better understand these factors at nano scale composites, we get a direct comparison between four polymers: poly methyl methacrylate (PMMA), polystyrene (PS), polyimide (PI), and poly-4-vinylpyridine (P4VP) with about half content (50% v/v) colloidal silica. Our data show that the electrical breakdown values through pure PMMA and pure PI are strongly reduced by adding even small quantities of highly dispersed colloidal silica (1% v/v). While, electrical breakdown values through Nano composites with low dielectric strength such as PS and P4VP increases with colloidal silica adding up to 15% v/v. In brief, cheap polymer Nano composites can easily develop amorphous polymers with low breakdown values especially if there are accompanied properties as flammability suppression and heat deflection.

Experimental

Silica nano composite films have been prepared using the same technique as described elsewhere [37–39] chemicals from Sigma–Aldrich: 1-poly methyl methacrylate PMMA, 2-methyl ethylketone MEK, 3-celite, 4-methanol, 5-polystyrene, 6-poly-4-vinylpyridine P4VP, 7-polyimide PI derived from pyromellitic dianhydride/oxydianiline PMDA/ODA. The obtained colloidal silica particles have radius about 15 nm with dispersion into the polystyrene and polyimide host matrices. Phenyl group density has been estimated using exclusion-chromatography techniques and the colloidal silica has been treated with phenyltrimethoxysilane to ensure homogenous dispersion through the polystyrene. If the dispersion finds a column, the unattached capping agents move through the column after the capped silica. Ultra violet measurements have been used in order to estimate the relative quantities of attached and unattached capping agents of the eluent which has been compared to the initial quantity of capping agent in the reaction to estimate graft density. The hydroxyl area of the uncapped silica has been effectively used with the P4VP and PMMA [40]. For about three days, the colloidal silica dispersion in di-methyl-formamide has been mixed with 10 wt% polymer solution in di-methyl-formamide which finally gives films of about 7 μm thick. Using 500 nm film of aluminum as a counter electrical contact, one ensures the building of a capacitor structure which gives a total thickness of about 7 μm . The electric breakdown and the dielectric characterization have been carried out using power supply that ensures about 10^4 V which is combined with a timing circuit that creates three hundred volts per second. Thus, the EBD time is about twenty seconds which is in harmony with previously published data [40]. The polymer nano composite film has been kept in direct attachment with a small copper hemispherical-ends-bar to give a direct electrical contacts with an effective contact area about 0.1 cm^2 . This makes good electrical contact and ensures removing the film heterogeneity due to initial manufacturing by local electric fields. The heads of bars are polished after each fifteen EBD. These 15 trails have been carried out for each film to calculate the most probable value of the particle size using Weibull distributions. To get continuous calibrated measurements, one has periodically used free-standing BOPP film to play the role of test-standard. Spellman generator with 10^4 V, Keithley 6517B and 1260A Solartron have been used to perform the electrical features of the composites. The time duration for each sweep has been kept at 30 s and

measurements are made at room temperature. The temperature variations at EBD due to Joule heating have been neglected. The measured leakage currents have been taken at half of the critical voltage of EBD.

Results and discussion

At 10^5 Hz, the above mentioned nano polymers have nearly the same dielectric constant; however, they markedly differ in the EBD voltages which are ranged from 4×10^8 V/m for PS to 8×10^8 V/m for PMMA. This shows how much polymer-colloid interactions are crucial to alter the EBD values. The continuous accumulation of nano particles is markedly embedded when using polar non-aqueous solvents (e.g. dimethyl formamide DMF and dimethylacetamide DMAC) when preparing the nano film [41] which enables creation of electric charges stabilization of physical properties of colloidal silica. To get an efficient drying rate, the polymer films have been coated onto a 100C-hot glass plate which embedded also the aggregation of nano particles. More time permits the solvent to evaporate so that the stability of electrical charges on the colloidal silica is highly reduced and the matrix viscosity becomes stronger enough to resist diffusion of particles which leads to strong dispersion of PNCs.

It is noted that this approach gives “qualitatively” similar particles of silica for all the composites even with high silica content (about 15% v/v) (see Fig. 1). At 10^5 Hz, the real and imaginary parts of complex permittivity ($\epsilon^* = \epsilon' + i\epsilon''$; with $i = \sqrt{-1}$) are plotted as a function of silica concentration (Fig. 2): in Fig. 2a, the dielectric constant. The dielectric constant ϵ' is plotted as a function of silica concentration and in Fig. 2b, the electrical conductivity σ is plotted as a function of silica concentration. Pure (0% v/v) films have the following dielectric constant: 2.6 for PS, 2.8 for PMMA, 2.9 for PI and 3.3 for P4VP which are in fair accordance with previously published study [42].

Fig. 2a shows the relative dielectric constant ϵ' of pure silica as a function of the applied frequency. This figure reveals that silica has high dielectric function. The dielectric constant of the presented films is illustrated in Fig. 2a as a function of the SiO_2 content. From this figure, one can see that ϵ' of the films raise linearly with the

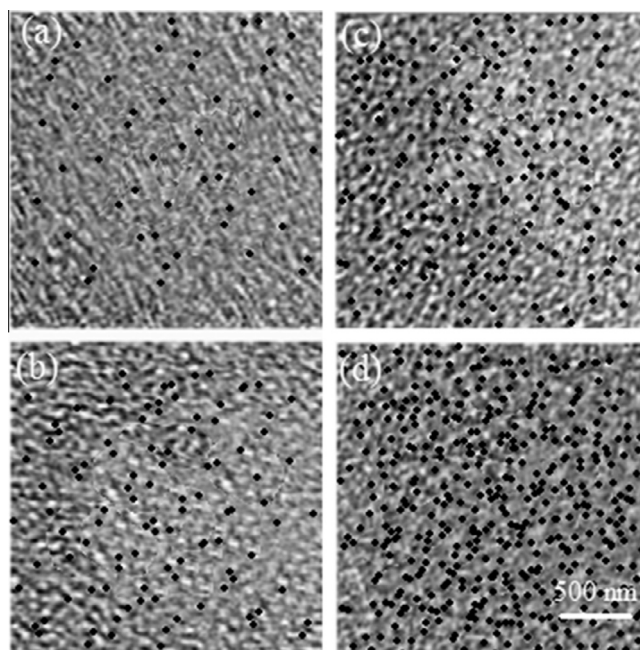


Fig. 1. TEM illustrations of polymer nano composite polystyrene films at different silica loadings: (a) 1%, (b) 5%, (c) 7.5%, and (d) 15%.

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