Layer-by-layer printed dielectrics for next-generation film capacitors for energy efficient space systems

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Abstract

Polymer film capacitors, composed typically of winding elements of thin metallized polypropylene films, are widely used in power electronics as DC-link capacitors for intermediate energy storage. However, the low energy density of the dielectric materials used in these capacitors can be a limiting factor, particularly in applications like spacecraft and space missions, where size and volume constraints are critical due to the high costs associated with launching objects into space. Polypropylene-based film capacitors have a specific energy density of 60-75 J/kg, which is significantly lower than that of electrochemical and ceramic energy storage devices. This limitation can potentially be addressed by incorporating ceramic nanoparticles into the polymer film. Our work has shown that, with the right surface treatment and choice of manufacturing technique, significant gains in specific capacitance and energy density can be achieved. In space applications, however, radiation sensitivity is also a key consideration. Our work shows that helium ion irradiation on polypropylene films reduces the thickness of polypropylene films, altering the capacitance. Our recent investigation showed that the thickness reduction of the irradiated material depends on the differences in the structure of both the metal coating and of the PP films, among other factors. While this radiation sensitivity is potentially detrimental to space applications, the thickness tuning effect may prove useful as a pre-treatment for the devices and can inform the type and scale of radiation shielding needed. Achieving these improvements with the melt extrusion method, typically used to manufacture film capacitors, is challenging – particularly if nanomaterials are to be used. We demonstrate a novel manufacturing method based on layer-by-layer printing. This approach allows for precise control over the thickness and composition of the dielectric film, achieving thicknesses well below 1 micron.

1. Introduction

A record number of objects (2664) were put into space in 2023 – two times more than in 2020 and twenty-two times more than in 2010.[1] That includes satellites for TV, radio, broadband, earth observation, ground navigation, as well as probes, landers, and space station flight elements, among others.[2] In space systems, power electronics play a critical role, as it is responsible for the efficient conversion, control, and distribution of electrical power to various systems onboard spacecraft, which might include power converters and voltage regulators.[3] Although the price of a space launch to low Earth orbit has dropped significantly within the last decades from 5400 USD per kg in 1960s (Saturn V, NASA) to below 2000 USD per kg currently (Falcon Heavy, SpaceX),[4] the size and weight of power

systems is a limiting factor for some types of satellites. Power systems can take up 18% of the mass or 20% of volume in the case of a nanosatellite.[5], [6] The size, volume and weight of power electronic devices depends on the required passive components, such us capacitors, which may occupy more than 25% of the entire system.[7], [8] Moreover, capacitors may take up a considerable volume in Pulsed Plasma Thrusters (PPT) that create high-power pulses used for spacecraft manoeuvring.[9] Capacitors are often used to smooth out voltage fluctuations and filter noise, thus help maintaining stable voltage for sensitive electronic equipment,[10] provide a buffer for energy collected from solar panels[11], [12] or thermoelectric generators.[13] Other applications such as collection of electrostatic charge from lunar regolith, and converting it into DC pulses through sequential charging and discharging episodes also exist.[14] Moreover, a capacitor-based electrical system in a nanosatellite have shown the potential for better durability, longevity, cost/volume efficiency, and lower charging time than standard lithium-ion batteries.[15] Therefore, improvements in the efficiency of capacitors can significantly affect a wide range of space systems, and for that application capacitors must have high energy density, stability over a wide temperature range, and resistance to radiation.

Film capacitors are the most frequently used and are responsible for 50% of the entire power capacitor market.[16] They incorporate a thin plastic film as the dielectric in between two electrodes, with variants including polyester and polypropylene, among many others. Their properties can be thermally stable within the -196°C-200°C (depending on the polymer used), while devices are ultra reliable via their self-healing capability, which is of paramount importance for space missions.[17], [18] Temperatures on the Moon can fluctuate between -180°C and +130°C, while the temperatures in shadowed areas near the lunar poles can fall as low as -230°C.[19] These temperature ranges make it difficult to use other types of capacitors. The electrolyte in electrolytic capacitors may become more viscous or freeze at low temperatures, which can increase the Equivalent Series Resistance or lead to a failure of the capacitor.[20], [21] Ceramic capacitors, on the other hand, can lose up to 95% of their capacitance at cryogenic temperatures,[22] and mechanical cracks in their structure might remain undetected during the mission.[23]

However, most polymers have dielectric constants below 3 [24], [25] and energy density of a polypropylene film capacitors is within 1-2 J/cm^3.[26] This is significantly lower than aluminium electrolytic capacitors (5.2 J/cm^3),[27] or other electrochemical energy storage devices.[28] Therefore, achieving high capacitance requires large volume and weight of capacitors and additional cooling systems, which is detrimental to volume and weight efficiency of power electronic systems. Improvements in storage capabilities of film capacitors can be achieved through incorporation of nanoparticles of high dielectric constant ceramics into polymers or blending linear and ferroelectric polymers.[29] However, polymer films are generally manufactured by melt extrusion, in which polymer pellets are sequentially heated and homogenized with a rotating screw, extruded, solidified and stretched to orient the polymer chains and reduced the thickness of the extruded films. The main advantage of this method is the mass production capability.[30] The addition of nanoparticles may lead to voids, cracks, and agglomeration during extrusion, which practically limits the weight fraction of the filler.[31], [32] On the other hand, alternative processing methods, such as solution

casting, can omit these issues but their production volume require upscaling for industrial usage.[33]

Recent work from our group has demonstrated that polymer nanocomposite film capacitors can be effectively produced using a novel gel-based layer-by-layer printing technique. This method leverages inks formulated with polypropylene and various industry-friendly solvents, which are deposited onto a conductive substrate. The novel approach not only allows precise control over the composition and thickness of the dielectric layers but also results in capacitors whose performance is on par with, if not superior to, commercial counterparts.[34], [35], [36] This paper focuses on the weight-reduction potential this method offers for power electronics components, exploring how the reduced mass can enhance efficiency, performance, and design flexibility. Additionally, the study examines the scalability of the technique for large-scale manufacturing and its implications for the future of lightweight, high-performance electronic devices. The ability to fine-tune material properties at the nanoscale opens new possibilities for custom-tailored electronic components, paving the way for innovations in various applications, including aerospace, automotive, and portable electronics.

2. Layer-by-layer printed dielectrics

Dispersing polypropylene in toluene and xylene at elevated temperature creates a gel that is stable at room temperature and can be deposited on top of a desired substrate by spin-coating (see Figure 1a). The method for this is detailed in a previous publication.[34] Due to low solubility and solvent resistance of polypropylene, it is possible to deposit successive layers of the gel without damaging the previous layers. This allows control of the thickness of the layers, which can be as low as 200 nm. The thin film may be made of a pure polymer or a nanocomposite with various weight fractions of ceramic nanoparticles. After deposition of the top electrode (see Figure 1d), the sample acts as a dielectric capacitor. The addition of nanofillers can significantly improve the capacitance and dielectric constant of the dielectric over neat polypropylene, but without precautions, this can also result in a reduction in breakdown voltage. This is most likely caused by defects in the devices which act as breakdown sites, which likely originate from nanoparticle clustering.[37] This is critical with respect to energy density.[32], [38] Volumetric energy density (U_v) is found from the expression in equation 1:

$$U_v = \frac{1}{2}\varepsilon_0\varepsilon_r E_{max}^2 \tag{1}$$

While specific energy density (U_m) is found from the expression in equation 2:

$$U_m = \frac{U_v}{\rho} = \frac{1}{2\rho}\varepsilon_0\varepsilon_r E_{max}^2 \tag{2}$$

Where ρ is the density of the dielectric, ε_0 is the vacuum permittivity, ε_r is the dielectric constant, and E_{max} is the breakdown strength (breakdown voltage per unit of dielectric thickness) of the dielectric. It is therefore clear from equations 1 and 2 that any impact on the breakdown voltage of the device will have a far greater impact on energy density than

any a similar change in the dielectric constant. Avoiding clustering is, therefore, the critical consideration in achieving high energy density nanocomposite dielectrics.

It was shown in our work that top-performing nanocomposite capacitors can have equal or higher breakdown voltages than neat polypropylene. This was demonstrated in theoretical analyses,[37], [39], [40], in preliminary device studies, [34], [36], [41], and in an upcoming comprehensive study of nanoparticle surface chemistry. Thus, the optimization of the weight fraction and surface chemistry of nanoparticles can reduce clustering and improve the overall performance of the nanocomposite film capacitors - particularly increasing the energy density. The possible chemical functionalization approaches include hydroxylation of the nanoparticles, the addition of surfactants, attachment of organic molecules, grafting of polymer chains or growing polymers directly from the nanoparticle's surface. Importantly, these chemical approaches are easier to apply to solvent-based nanoparticle dispersions (such as polypropylene gels) than to melt extruded films, which require high temperature and tedious homogenization steps. However, so far, printed dielectrics have only been successfully manufactured in laboratory scales, and it remains to be demonstrated that these materials can be fabricated reliably using industrial-scale fabrication techniques. One promising way towards large-scale production is slot-die coating, which is an established manufacturing methods for photovoltaics and could potentially be extended for layer-by-layer fabrication of nanocomposite capacitor devices using polymer-based inks.[34], [35], [36], [42]

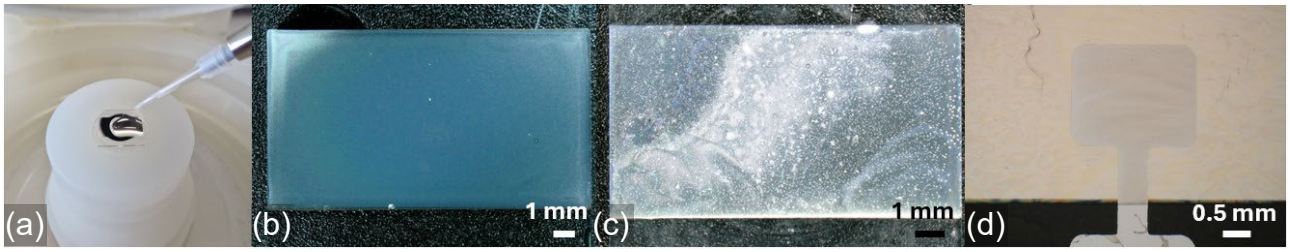

Figure 1. Spin coating of polypropylene gel onto a conductive substrate (a), a polymer dielectric (b), nanocomposite dielectric layers (c), and a top aluminium electrode deposited onto the dielectric layer (d).

3. Dielectric properties

Commercial biaxially oriented polypropylene (BOPP) films typically have a dielectric constant of 2.2-2.3 and films with a thickness of 5 μm or 10 μm exhibit breakdown strength of up to 727 V μm^{-1} and 811 V μm^{-1} at room temperature, respectively.[43] On the other hand, the PP capacitors manufactured with our process have a thickness of 700 nm and an unoptimised breakdown strength of 350 V μm^{-1} [31] (this figure will be improved upon in upcomming publications). The addition of 5 wt.% (or 0.8 vol.%) of barium titanate nanoparticles into the gel decreased the breakdown strength to 300 V μm^{-1} and the mass increased by 8%, while the capacitance increased by 12%.

It is worth noting that all benefits we see in dielectric performance occur at low volume fractions of nanoparticles, where the additional mass of the nanoparticles has a comparatively small effect on the density of the dielectric, as described in equation 3.

$$\frac{1}{\rho_{comp}} = \frac{x_{NP}}{\rho_{NP}} + \frac{1 - x_{NP}}{\rho_{poly}} \tag{3}$$

Where ρ_α is the density of the nanocomposite, the nanoparticle material, and the polymer matrix material for $\alpha = comp, NP, poly$ respectively, and x_{NP} is the wight fraction of nanoparticles in the nanocomposite. For very small values of x_{NP}, it follows that the density of the composite (ρ_{comp}) will vary little from the density of the polymer material (ρ_{poly}), and therefore have minimal impact on specific energy density (cf. equation 2).

Our recent work has shown that the surface functionalization of nanoparticles can lead to devices with simultaneously improved capacitance and breakdown strength. The addition of an ionic surfactant to the PP gel containing nanoparticles resulted in devices that had up to 39% higher capacitance, while a non-ionic surfactant increased breakdown voltage by 70%.[41] For example, a pulsed plasma thruster ADD SIMP-LEX (ADvanceD Stuttgart Impulsing MagnetoPlasmadynamic thruster for Lunar Exploration) is composed of 4 capacitors with a capacitance of 20 μF and a maximum voltage of 1300 V and a maximum stored energy of 68 J.[44] Thus, the capacitors would weigh 0.2 kg if made of the printed PP, but only 0.1 kg if the dielectric was replaced with the composite with nanoparticles. The main advantage of the printed dielectrics is their much higher specific capacitance than the extruded BOPP ($9 \cdot 10^{-4}$ F/kg). The printed PP devices exhibit 51 times higher specific capacitance, while the nanocomposite 52 times higher. In this way, the method has a potential to significantly reduce the size and weight of power electronic components.

4. Helium irradiation

We have carried out helium ion irradiation experiments with our devices, primarily as a fundamental investigation of the effects that such irradiation has on polymer and polymer nanocomposite films, but the results of this have relevance for space applications as well due to the similarity to the charged particle radiation environment in Earth orbit and in deep space.

Helium ion irradiation of metallized polypropylene (PP) modifies its bulk by structural changes in the polymer chains, while preserving the metal layer. Irradiation of polymer with helium ions can result in the breaking of the chemical bonds in the polymer chains, cross-linking, forming free radicals and double bonds among others changes.[38], [45] We irradiated both spin-coated PP and extruded BOPP metallized films with He^+ ions and studied the effects of irradiation on the surface and the bulk (sub-surface) of PP. The irradiation was done with a He^+-FIB in a Zeiss Orion NanoFab Microscope at a landing energy of 25 keV with doses ranging from 5.4×10^{-5} nC/μm^2 to 8.07×10^{-3} nC/μm^2.[46] He-FIB was used to construct surface patterns similar to those fabricated in our previous studies

[46], [47], [48] of other polymer materials irradiated similarly. Atomic force microscopy (AFM), Raman spectroscopy, and optical microscopy were used to analyse the details of surface modification. He^+-ion irradiation reduces, in a controlled fashion, the surface height of the irradiated regions.[46] Three metal coatings: Au, $Pt_{60}Pd_{40}$, and Al were separately applied on the PP film surface prior to irradiation. Among them, the Al-coated samples demonstrated smaller surface-height reduction as compared to the $Pt_{60}Pd_{40}$ and Au-coated samples. The possible factors responsible for this effect include differences in the thickness and the crystalline-grain orientation (texture) of the metallization films. The shape and size of the irradiated area do not alter the irradiation effect, as shown in Figure 2. Preliminary results indicate an increase in the capacitance and the breakdown voltage after He^+-ion irradiation on PP-based metallized film capacitors. The effects on nanocomposite films will be discussed in an upcoming publication.

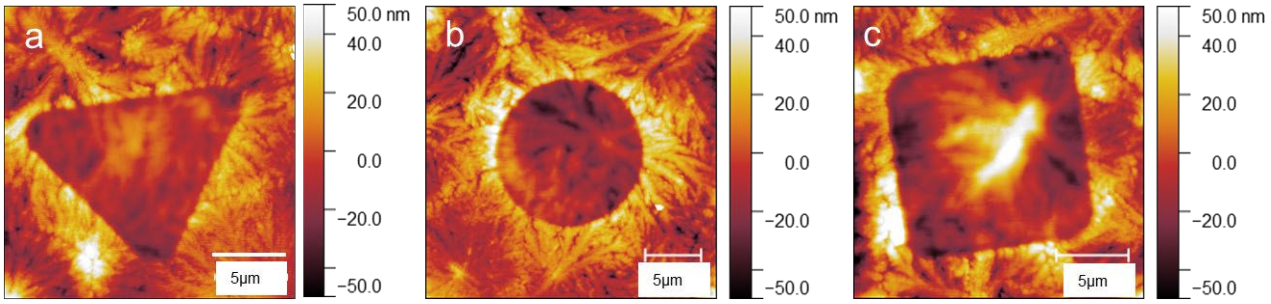

Figure 2. Different shaped a) Triangular, b) Round, and c) Square-shaped AFM images of the spin-coated PP film after He^+-FIB irradiation with the dose $5.4x10^{-5}$ nC/µm^2

5. Conclusions

Nanocomposite dielectrics are a promising material class emerging in capacitor research as a way to bridge the gap between the high capacitance but low reliability and voltage tolerance of ceramic dielectrics, and the high breakdown strength and good reliability but poor volumetric and specific capacitance of polymers. Our work has shown a viable and scalable way to fabricate nanocomposite dielectric capacitors that minimises clustering, ensuring maximal energy density is obtained. By combining this method with nanoparticle surface chemistry engineering, it is possible to realise significant gains in specific capacitive energy density, which could lead to substantial reductions in the weight of satellite power electronics systems if implemented in such an application.

We have also carried out preliminary studies of the effects of helium-ion irradiation on polypropylene films produced from polymer-gel inks, demonstrating that this radiation can alter the capacitance of the material. Such alterations would be problematic if they occurred in an installed system, however the presence of such effects is not unique to polymer capacitors and may not significantly alter the radiation shielding requirements of the devices (although a longer-term reliability study would be needed to confirm this). Where these results may also prove useful is by incorporating an He-ion pre-treatment into the manufacturing process to further improve the device properties, although such a process

would have to take into account any potential interaction between the nanoparticles and the He-ion radiation.

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