Nanocomposite Film Dielectrics for High Temperature Power Conditioning Capacitors

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Abstract

Emerging power electronics in a broad range of military, aerospace, hybrid vehicle, renewable energy and drilling applications rely on advances in dielectric materials for capacitors. Compact capacitors possessing low dielectric loss and high operating temperature capability are needed for power conditioning in advanced converter and inverter designs for these applications. Wound film capacitors represent the preferred capacitor technology but are limited by low operating temperature capabilities, < 150°C, and low volumetric efficiency, capacitance per unit volume less than 1.0 µF/cc.

TPL is developing processes and manufacturing techniques for fabrication of advanced capacitor films comprised of high temperature polymers modified with ceramic nanoparticles. The selected polymers enable high temperature operation and the ceramic nanoparticles enhance volumetric efficiency by increasing the dielectric constant. The fabricated films are directly adaptable to conventional wound film capacitor construction methodologies.

This paper provides an overview of TPL's experience to-date with a film dielectric material system comprised of fluorenone polyester polymer and titanate nanoparticle filler. The fluorenone polyester polymer was selected to provide a stable operating temperature of at least 300°C, while the addition of the titanate powder increases the dielectric constant and, in turn, the volumetric efficiency of wound film capacitors constructed with this film system. Status and ongoing capacitor development issues are provided relative to the intended high temperature power conditioning applications.

Key Words: fluorenone polyester, nanocomposite, capacitor, volumetric efficiency

General Issues Surrounding High Temperature Capacitor Films

Various high temperature polymers are available today such as polyimide, silicone, polyethersulfone and polytetrafluoroethylene but have not been manufactured into capacitor film required to achieve high volumetric efficiency. (Limitations on process-ability for capacitor film are unique to each polymer and are beyond the scope of this paper.) One exception is fluorenone polyester (FPE), a polymer originally developed by 3M in the 1980s. FPE provides the physical properties, process-ability and thermal properties required for the fabrication of capacitors capable of operating at temperatures in excess of 300°C. Recent work funded by the US Department of Energy supports progress toward this development of FPE capacitors [1].

While capacitors with increased operating temperature capability appear attainable, achieving increased volumetric efficiency is uncertain with current polymer technology. Limits to the parameters controlling volumetric efficiency, as described below, are largely defined.

$$\text{Volumetric Efficiency} \propto [\text{Ů}t] \times PF$$

where: Ů= dielectric constant
t = capacitor film thickness
PF = capacitor packaging factor
(active/total volume)

The dielectric constant of all low dielectric loss polymers falls within a narrow range, 2.5 to 3.5. Capability for producing and processing capacitor film is limited to a narrow thickness range, 2.0 µm to 5.0 µm. Packaging efficiency of dielectrics in wound film capacitors typically falls within a narrow range, 0.65 to 0.75.

TPLôs approach to producing high temperature capacitors with high volumetric efficiency involves modifying polymers to achieve an increased dielectric constant. Thermally stable ceramic dielectric fillers possessing a high dielectric constant (k > 1,000) are uniformly dispersed in the polymer to form a composite (Figure 1). Incorporation of sub-micron titanate particles have been used to increase the polymer dielectric constant by a factor 20, e.g., 3.0 to 60. In the case of capacitor film, a filler concentration is used that does not significantly impact the mechanical properties, dielectric loss or voltage stress capability. Thin capacitor films, 2.0 µm to 5.0 µm, can be produced with a three to five-fold increase in dielectric constant, relative to the unmodified polymer. This increase in dielectric constant is directly proportion to increased volumetric efficiency.

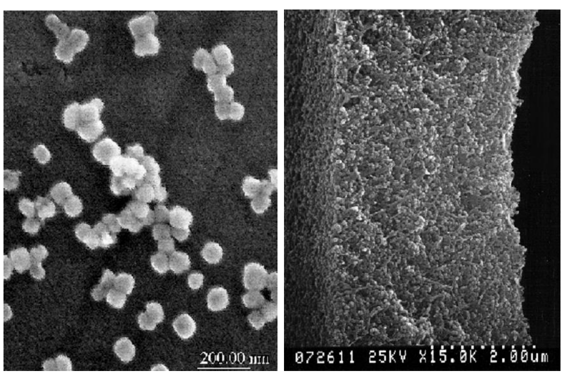


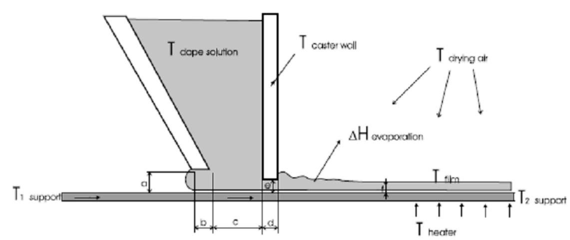
Figure 1: Micrograph of TPL titanate particles (50 nm) and composite dielectric film (4 µm).

Processing of the dielectric film into highly efficient and reliable capacitor designs is achieved in wound configurations that provide tolerance to shorting due to dielectric failure at voltage. Self-healing, or soft breakdown, behavior represents a major reliability advantage that metallized-film capacitors possess. It derives from the thin metal layer (measured in angstroms) of metal that is applied to the film to form the capacitor plates. When a dielectric breakdown occurs, electrical charge flows rapidly to the breakdown site. This large surge of current vaporizes the metal around the breakdown site. Removal of the metal around the breakdown site isolates the breakdown site from the voltage stress and, thus, the capacitor self-heals. In contrast, typical ceramic capacitors have relatively thick electrodes that are printed from metal particle dispersions and co-fired with the ceramic; accordingly, there is no self-healing behavior.

Through a combination of ceramic processing and solvent-based coating processes, TPL has engineered composite dielectric films that possess the electrical and physical properties required for producing capacitors with a substantial increase in operating temperature and volumetric efficiency. TPL has investigated capability for producing capacitor grade composite films of several high temperature polymers including polyimide, silicones, epoxies, polyetherimide, polysulfone, polyethersulfone and fluorenone polyester (FPE). FPE nanocomposite has been pursued to the largest extent to-date and is, therefore, selected for this description of development and fabrication aspects. FPE itself provides a stable operating temperature of at least 300°C. As described in this paper, titanate nanoparticle modifications to FPE can provide a factor of three increase in capacitor volumetric efficiency while maintaining temperature capability of at least 300°C.

Film Fabrication Methodology

The film fabrication methodology utilized by TPL involves casting of the nanoparticle-polymer dispersion onto a supporting and releasable substrate using a doctor blade casting head (Figure 2), and in-line ovens to remove solvents from the cast solution (Figure 3). The result is nanocomposite film on a releasable substrate (Figure 4).

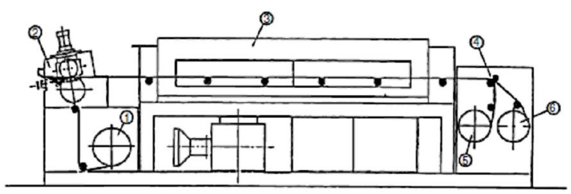


a. ï f. coating head dimensions
T temperatures
H enthalpy of solvent(s)

Figure 2: Doctor blade casting head [2]

TPL has investigated several techniques for fabrication of capacitor-grade nanocomposite films in the thickness range of 2 µm to 15 µm. Given the ability to solvate the polymer resin in the nanocomposite system, film fabrication by doctor blade coating on an ultra-smooth substrate has been found to provide the best and most consistent results. Other techniques investigated include slot die, rod and gravure coating; however, each of these alternate techniques was found to have at least one aspect that precluded quality film formation with acceptable thickness uniformity in a production process.

Separation of the film from the substrate can be accomplished as a part of the in-line casting and drying operation, as depicted by the support film and product film winders in Figure 3. Alternatively, TPL has developed methods for separation of the film and substrate during the capacitor winding operations, Figure 4. Separation at winding enables especially delicate films to be conveyed through intermediate operations such as metallization, slitting and spooling with reduced susceptibility to film damage.



1 = support film un-winder
2 = casting head (doctor blade)
3 = flat bed dryer
4 = separation point
5 = support film winder
6 = product film winder

Figure 3: Cross-section of film casting line [2]

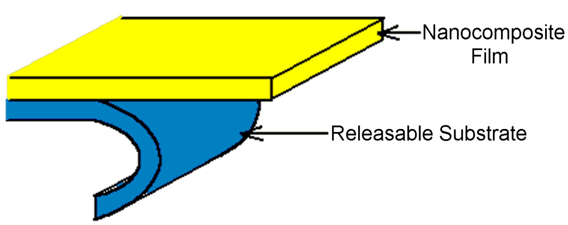


Figure 4: Nanocomposite film cast onto supporting, releasable substrate

Composite Dielectric and Film Development

Initial material and process development involved spin coating and a laboratory-scale casting line to investigate all fundamental formulation and processing parameters. Emphasis was placed on fabricating films at less than or equal to 4.0 µm with adequate mechanical properties for removal from the support substrate and routine film evaluation activities. Defining a material formulation and set of process parameters that provided an increase in film dielectric constant without impact on other critical performance characteristics was a primary objective. The performance objective was a capacitor film at a thickness less than 4.0 µm capable of operating at a voltage stress of 200 V/µm and greater.

Sheet capacitor testing (up to 30 cm^2) was used to establish the basic nanocomposite system composition, solution conditions and dielectric properties. Provided a baseline system established with spin coating, film casting was then transferred to the laboratory-scale casting line for continued analysis and refinement. A primary focus of this effort was to achieve high quality film that provided reliable performance under voltage stress. Conditions and processes were defined for fabricating FPE nanocomposite film at the target thickness. Several coating trials allowed for the determination of the proper casting rate, temperatures and deposition conditions to achieve high quality film with suitable mechanical properties at thicknesses down to 3.5 µm.

Laboratory-scale casting trials facilitated identification of the critical formulation and process parameters, and establishment of guidelines for transference of casting conditions TPLôs recently established pilot-production coater. Film thickness and uniformity are controlled rheologically through solvent selection, polymer solution viscosity and polymer concentration in solution, and physically through film feed rate and die head adjustments. Time and temperature profiles and exhaust rates also play a major role in the drying rate as the film is conveyed through a series of oven zones.

Pilot-Production Scale Film and Capacitor Development

Provided success in achieving target film performance on the laboratory-scale coater, film fabrication was transitioned to a government-furnished-equipment pilot-scale coating line. This pilot-scale line was installed in October 2010 and became fully operational in mid-2011. Procurement of this line was supported by the U.S Navy, Office of Naval Research, for pilot-scale fabrication of advanced dielectric films under development by TPL and other organizations receiving U.S. Department of Defense support. The line has 3 independent oven zones, a nominal line rate of 90 meters per hour, a maximum coated width of 375 mm and is housed in a Class 100 clean room, Figures 5 and 6.

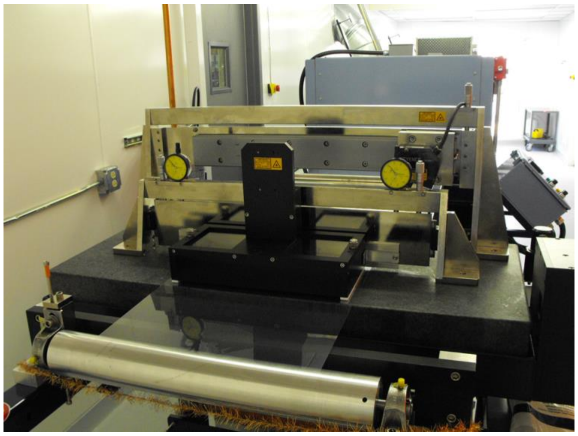


Figure 5: Pilot-scale solvent-cast film fabrication line, coating head area

Figure 6: Pilot-scale solvent-cast film fabrication line, oven exit area

At least 100 m^2 of continuous quality film is required for the film processes necessary to support prototype capacitor fabrication. This line provides such capability as well as a clean environment to minimize particulate inclusions that can significantly degrade the filmôs voltage reliability. All major film fabrication work on the FPE nanocomposite has been successfully transitioned to this pilot-scale line.

A significant number of film production trials focused on optimizing coating parameters have led to film quality that supports consistent capacitor performance. Film properties are consistent with previously reported data [3]. Concurrently, aspects related to capacitor manufacturing using the composite film have been established. To-date, all process parameters required for fabricating FPE nanocomposite capacitors have been successful addressed. Figure 7 shows the critical process stages required to convert the FPE from the nanoparticle dispersion to the final capacitor section. Film rolls of the FPE composite were metallized, slit and spooled into roll pairs suitable for capacitor winding, Figure 8. Provided metallized film in roll stock, capacitor sections were wound and terminated, Figure 9.

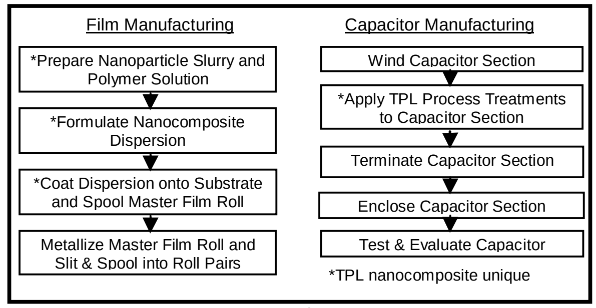


Figure 7: Process stages for FPE nanocomposite capacitor fabrication.

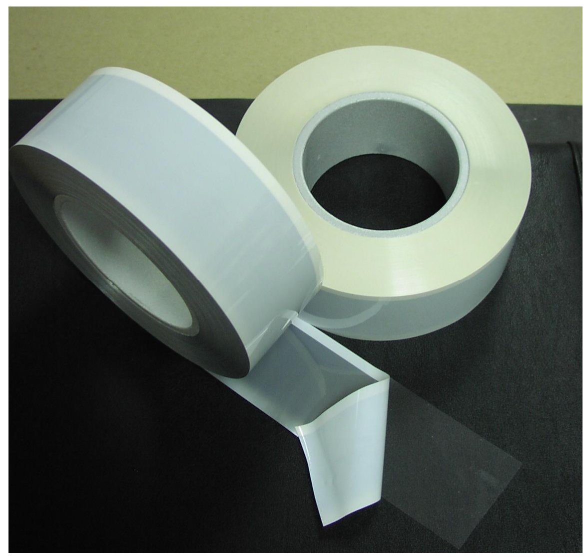


Figure 8: Metallized, slit and spooled rolls of FPE nanocomposite film ready for capacitor winding.

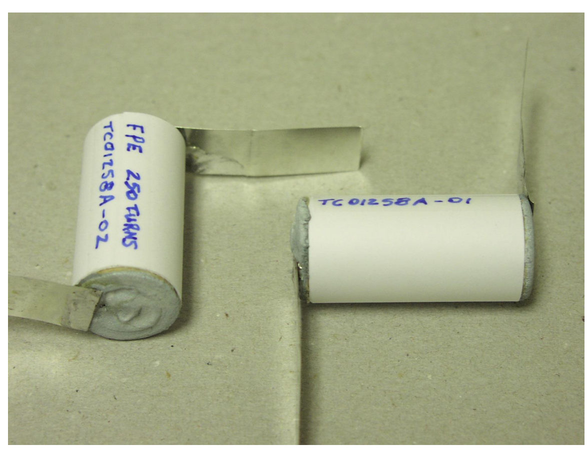


Figure 9: Wound and terminated FPE nanocomposite capacitor sections

An encapsulating material for the capacitors was recently established and offers a unique packaging solution. The encapsulant is a high temperature elastomer modified with ceramic nanoparticles (Figure 10) which helps to reduce thermal expansion and permeability, increase dielectric constant, and increase thermal conductivity. The unique capacitor enclosure provides: (1) environmental protection for the capacitor winding and terminations; (2) operational temperature capability from -55 to +300 ºC; (3) electrical interconnection and heat transfer interfacing to the targeted DC-DC converter system; (4) minimal addition to overall capacitor volume.

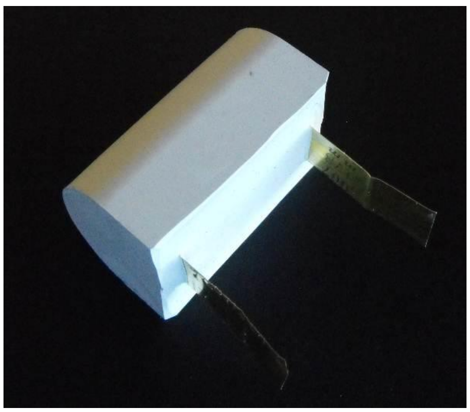


Figure 10: Encapsulated FPE nanocomposite capacitor

Recent Development Focus for Capacitor Operation at 200+ °C

Ongoing development activities are focused on addressing specific material and design aspects relative to application requirements. Prototype capacitors (5 to 10 µF) are being routinely fabricated with 4.0 µm FPE nanocomposite film. Testing of these capacitors supports a largely independent temperature and frequency behavior with less than ± 5% change in capacitance from -55 to +200 °C and 100 Hz to 100 kHz. Most recent capacitor testing has shifted to establishing performance under simulated application requirements (ripple testing). Operational limits of the capacitors are being assessed relative to current, voltage and temperature. Material and design aspects for further development are being identified based upon these test findings.

The basic ripple test configuration is a constant-charge, pulse discharge arrangement, as depicted in Figure 11. DUT denotes the capacitor being tested. Cooling and heating apparatus have been adapted to the test configuration to allow adjustment of the DUT temperature from -55 to +300 °C. Figure 12 is a representative oscilloscope image showing voltage and current waveforms during ripple testing of a 6.2 µF FPE nanocomposite capacitor at 600 V_{dc}, 5.0 A_{rms} and 150 kHz.

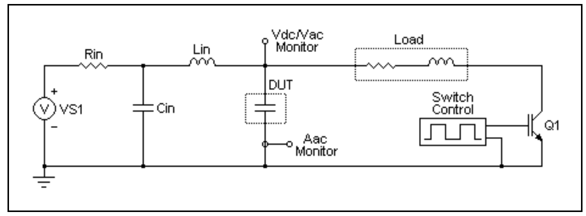


Figure 11: Capacitor ripple current test configuration

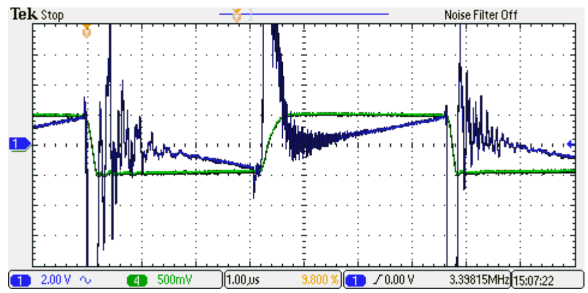


Figure 12: Ripple test voltage and current waveforms (voltage on Ch1, current on Ch4 @ 0.1V/A)

The critical capacitor performance finding to-date is good voltage and ripple current performance up to 200 °C. Recent results support capability operating at high ripple current and high voltage, 0.8 A/µF and 500 V respectively, at a switching frequency of 150 kHz. Above 200 °C, failure rates due to shorting are unacceptably high. This source of failure has been traced to excessive internal heating within the capacitor during ripple test operations. This heating is attributable to the equivalent series resistance (ESR) of the capacitor and dielectric loss of the FPE nanocomposite.

The capacitor ESR is primarily attributable to the electrode metallization resistivity and capacitor geometry (film length-width ratio). This is a conventional wound metallized film capacitor design issue wherein the electrode resistivity and capacitor geometry must be carefully selected to achieve a specific ESR and, thereby, ripple current capability. The unique aspect associated with FPE nanocomposite film is the increased surface charge per unit film area that results from its increased dielectric constant relative to conventional polymer films [4]. Reduced electrode resistivity levels and increased film length-width ratios are presently being prototyped and evaluated for the best balance of ESR (ripple current capability) and volumetric efficiency.

Relative to the heating associated with ESR, the dielectric loss of the FPE nanocomposite is much more subtle. The loss appears as a significant decrease in insulation resistance at internal capacitor temperatures above ~ 225 °C and, upon initiation, behaves in a self-generating manner. The typical result is relatively rapid failure at operating temperatures above 200 °C with no apparent measurable or visual indication of failure onset in the ripple test configuration.

Leakage current analyses were conducted on prototype capacitors as a function of temperature. These analyses revealed a pronounced inflection point between 200 and 225 °C with high rate of leakage current increase with increasing temperature thereafter. This testing supports the ripple test findings and thermal runaway when operating capacitors above 200 °C.

Recent refinements to the material processes, beyond the scope of this paper, have been successful in reducing the dielectric loss of the FPE nanocomposite. The method found most practical for timely analyses of the film loss is measurement of the 100 Hz dissipation factor. Figure 13 shows the 100 Hz dissipation of two different film samples measured over the range 30 ï 300 °C. The ñBaselineò film sample curve shows the 100 Hz dissipation behavior at the start of efforts to address this high temperature loss problem; note the relatively rapid increase in dissipation above 200 °C. The ñImprovedò film sample curve shows the 100 Hz dissipation behavior after initial steps were taken to reduce the concentration of charge carriers in the FPE nanocomposite. The result was an approximate 50% reduction in dissipation factor over the entire temperature range.

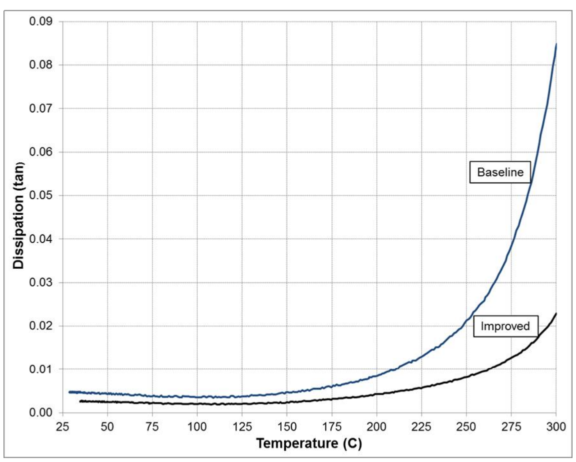


Figure 13: 100 Hz dissipation factor before and after first FPE nanocomposite process purification steps

Further refinements of the material processes are anticipated and are currently being incorporated in film for capacitor evaluation. These material improvements combined with a reduced capacitor ESR are expected to minimize the internal heating issue and allow for capacitor operation above 200 °C.

Conclusions

TPLôs work with FPE nanocomposite film to-date has demonstrated the potential for achieving compact, high temperature filter capacitors for emerging power conditioning applications.

The principle advancement over high temperature polymer films is an increase in dielectric constant by a factor of two through the addition of titanate nanoparticle fillers. The volumetric efficiency of the FPE nanocomposite film capacitors is projected to be greater than 1.0 $\mu F/cm^3$ for high ripple current operation at 400 to 600 Vdc.

To-date, prototype wound film capacitors demonstrate favorable temperature stability over the operating range of -55 to +200 °C and 100 Hz to 100 kHz. Capacitors demonstrate capability for high current and high voltage operation, 0.8 A/µF and 500 V respectively, at a switching frequency of 150 kHz.

Continued development activities directed at reducing capacitor losses that result internal heating are expected to result in an operating temperature capability greater than 200 °C.

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