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Dielectric breakdown studies of Teflon perfluoroalkoxy at high temperature

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Teflon® perfluoroalkoxy (PFA) was evaluated for use as a dielectric material in high-temperature high-voltage capacitors for space applications. The properties that were characterized included the d.c. dielectric strength at temperatures up to 250°C and the permittivity and dielectric loss as a function of frequency, temperature and voltage. To understand the breakdown mechanism taking place at high temperatures, the pre-breakdown discharge and conduction currents, and the dependence of dielectric strength on thickness of the film were determined. Confocal laser microscopy was performed to diagnose for microimperfections within the film structure. The results obtained show a significant decrease in the dielectric strength and an increase in dielectric loss with an increase in temperature, suggesting that impulse thermal breakdown could be a responsible mechanism in PFA film at temperatures above 150°C.

Introduction

As space power requirements move into the multi kilowatt range, electrical insulation and dielectric materials become the critical issue that needs to be given greater attention in order to provide higher reliabilities and energy densities in space power systems [1]. A major thrust of research in this area has been and will continue to be the development of new dielectric materials to increase the capability of energy transport and storage devices to withstand a higher level of space environmental stresses, including thermal and radiation [1].

To meet high space power demand, it might become necessary to use nuclear reactors, among other prime power sources, because they offer significantly higher energy densities and compactness [1]. A smaller volume of the space vehicle to minimize mass and thrust and lower efficiency of heat rejection in space will contribute greatly to an increase in the surrounding temperature [2]. Because the dielectric properties of polymeric materials change with temperature, the temperature dependence of their properties is of prime importance in analysing their breakdown mechanism. A single-point failure in the insulation and dielectric may very well prove catastrophic for the whole mission or part of the system, as was experienced in the *Voyager's* receiver circuit in the vicinity of higher levels of thermal stress over an extended period of time [3].

There have been numerous studies on the behaviour of dielectric materials, but they have been carried out at relatively lower temperatures, typically up to 90° C [4, 5]. This is because most dielectric materials, mainly polymeric, used for energy storage and transport devices have limited maximum service temperature. With the exception of Kapton® (polyimide), which is used as an insulator in space, very little has been investigated to explore the possibility of an alternate high-temperature polymer dielectric which can withstand considerable levels of high voltage. Polyimide is, however, reported to have a drastic reduction in breakdown strength at temperatures above 200° C [6], and has a maximum service temperature of 240° C [7]. Polyimide also suffers from significant reduction in volume resistivity above 100° C [8]. A literature search shows that perfluoroalkoxy (PFA) film, on the other hand, has a higher service temperature to 260° C and better thermal conductivity, mechanical properties and heat resistance compared to polyimide [9–11]. Very little information, however, is available on the high-voltage electrical characterization of PFA film at high temperatures, with the exception of earlier works by Gangal [12] and Bro *et al.*, [13].

In this paper, the high-temperature dielectric studies of Teflon® PFA film are reported. The dielectric properties measured include the d.c. breakdown voltage at temperatures up to 250° C, and the permittivity and dielectric loss as a function of frequency in the range 50 Hz–100 kHz, temperatures up to 200°C and electrical stresses up to 40 V μm^{-1}. To understand and explain the breakdown mechanism taking place at these higher temperatures, the partial discharge activity, pre-breakdown conduction currents and the dependence of dielectric strength on thickness of the film were determined. Confocal laser microscopy was also performed to diagnose the as-received PFA film for microimperfections, such as air cavities, within the film structure.

2. Experimental procedure

PFA films of 25 μm thickness (± 1 μm), were mostly

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used in these investigations. The reported electrical and physical properties of PFA material are given in Table I [9–11].

The properties that were measured in the present work include the permittivity and dielectric loss as a function of frequency, temperature and voltage and the d.c. breakdown voltage at temperatures up to 250 °C. The permittivity and dielectric loss were measured at room temperature using GenRad 1689 Precision RLC Digibridge at eight different frequencies ranging from 50 Hz–100 kHz. The details of the measurement technique and the electrode system are reported elsewhere [14]. The specimen surfaces were deposited with about 100 nm thick aluminium electrodes to ensure good contact for all dielectric measurements. The permittivity and dielectric loss measurements were also carried out at high voltages and high temperatures using a Tettex Instrument Precision Measuring System, Type 2822. These measurements were performed at temperatures up to 200° C at 200 V and 60 Hz. The permittivity and dielectric loss were also measured as a function of voltage up to 1000 V at room temperature using the Tettex System.

The breakdown voltages of the films were obtained by using a Hipotronics d.c. Corona Free Dielectric Test Set, Model 230-10CT. A bath of silicone fluid 210 H, a high-temperature dielectric fluid, was used with a temperature controller to obtain proper test temperatures ± 2° C. The experimental apparatus for breakdown voltage measurement is shown in Fig. 1. During testing, the film was placed between two cylindrical stainless steel electrodes of 2.54 cm diameter (ASTM D-149) and the voltage was increased at a rate of 500 V s^{-1} until breakdown occurred. A ceramic fixture was employed to support the electrodes. The values reported for breakdown are the averages of seven measurements with the standard deviation shown.

The partial discharge activities were monitored using a portable Partial Discharge Analyser (PDA), Biddle Instrument Co., Model-4, and the method described elsewhere [14]. The resolution of this PDA was less than 6 pC. Conduction currents were measured at voltage steps of 1 kV until breakdown occurred for various temperatures ranging from 22–250° C. The measurements were made after a 3 min time interval on application of each voltage step [15].

TABLE I Reported properties of PFA Film at 22° C[9–11]

Chemical Structure	$[(CF_2 - CF_2)_n - CF_2 - CF(OC_mF_{2n+1})]_m$
Maximum service temperature (°C)	260
Thermal conductivity (Wm^{-1}K^{-1})	0.19
Water absorption (%)	0.02
Elongation at break (%)	200–300
Tensile strength (N m^{-2})	1.38×10^7
Young's modulus (N m^{-2})	4.80×10^8
Dielectric strength V μm^{-1}	118–256
(25.4 μm and ASTM D-149) [kV mil^{-1}]	(3.0–6.5)
Permittivity (at 1 kHz)	2.15
Dielectric loss (at 1 kHz)	0.0002–0.0007
Volume resistivity, (Ωcm) at 22–240° C	$> 1 \times 10^{16}$
Density (g cm^{-3})	2.13

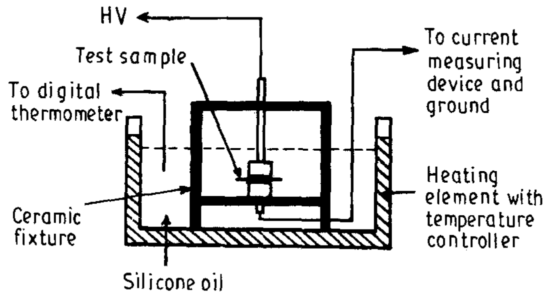

Figure 1 Experimental apparatus used for breakdown studies. HV = high voltage.

A BioRad MRC-500 Unit was used to carry out confocal laser microscopy on PFA samples. An argon-ion laser beam having a 25 mW power capability, and 488 and 514 nm wavelengths, was utilized to scan layers of the microstructure. The confocal microscope employed Nikon Fluor 40X lens with a numerical aperture of 1.3. The sample was placed on an aluminium slide-like fixture. The film surface to be scanned was covered with Immersionsoel ($n_e = 1.518$), to obtain sharp images of the microstructure.

3. Results

The permittivity and dielectric loss of PFA film as a function of frequency, ranging from 50 Hz–100 kHz, at room temperature are shown in Fig. 2. The permittivity of the PFA film is essentially independent of frequency. However, the dielectric loss was found to increase with frequency, especially above 1000 Hz. Fig. 3 shows the permittivity and dielectric loss of PFA film as a function of temperature. A slight reduction, though not very significant, in the permittivity is noticeable with an increase in temperature. However, the dielectric loss increases with temperature. The permittivity and dielectric loss as a function of voltage are shown in Fig. 4. There is no appreciable change in these properties at voltages up to 400 V. As the voltage is further raised, the dielectric loss shows an increase, particularly at 500 V.

The dependence of d.c. dielectric strength on temperature is shown in Fig. 5. The breakdown voltage of PFA film exhibits an almost independent relationship up to 150° C, but beyond this temperature, shows a considerable decrease with a rise in temperature. The absolute values of partial discharges are tabulated for several voltage levels, at room temperature, in Table II. The partial discharge level increases with applied voltage particularly above 400 V.

With a view towards understanding the failure mechanism taking place at higher temperatures, additional breakdown experiments using 51 and 127 μm thick (± 2 μm) PFA films were carried out at 22, 125 and 250° C to determine the thickness dependency of dielectric strength. The results are shown in Fig. 6. Each datum point represents an average of seven measurements. At room temperature, the breakdown strength decreases with increase in thickness. This thickness dependency becomes mild at 125° C and nil at 250° C. In support of thermal breakdown theory, Fig. 7 shows the conduction current density as a

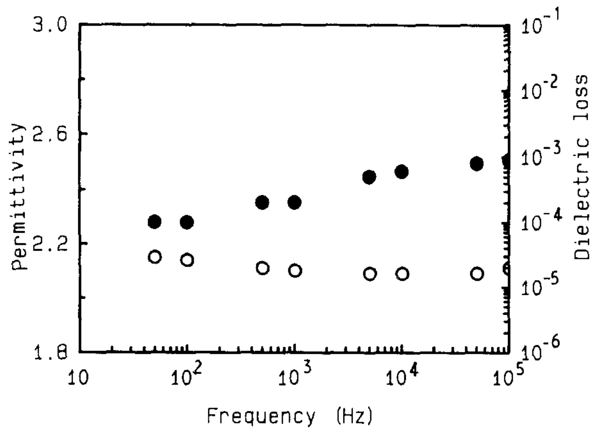

Figure 2 Effect of frequency on (○) relative permittivity and (●) dielectric loss of PFA film at 22°C.

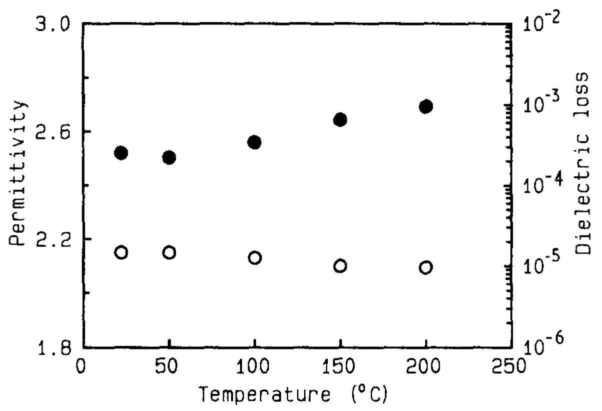

Figure 3 Temperature dependence on (○) relative permittivity and (●) dielectric loss of PFA film at 8 V μm^{-1} and 60 Hz.

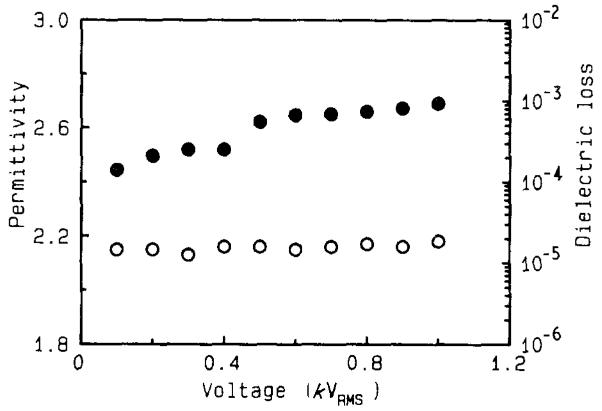

Figure 4 Voltage dependence on (○) relative permittivity and (●) dielectric loss of 25.4 μm thick PFA film at 22°C and 60 Hz.

function of voltages at different temperatures. The conduction current greatly increases with an increase in both voltage and particularly the temperature.

In order to examine the microstructure of PFA film, confocal laser microscopy was performed and the photographic image is shown in Fig. 8. The image was taken about 10 μm deep from the film surface for a virgin sample. Micro-voids of submicrometre size are visible within the film structure. A better resolution could not be obtained due to the limitation of the instrumentation used.

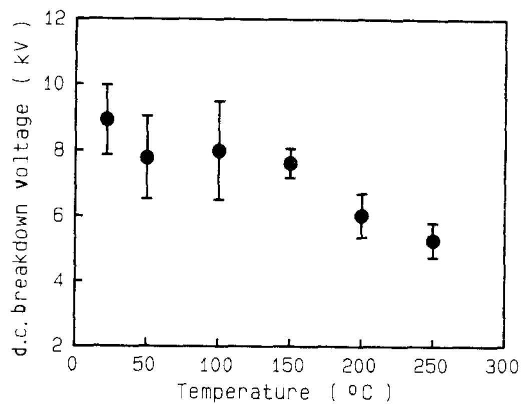

Figure 5 Temperature dependency of d.c. breakdown strength of PFA film.

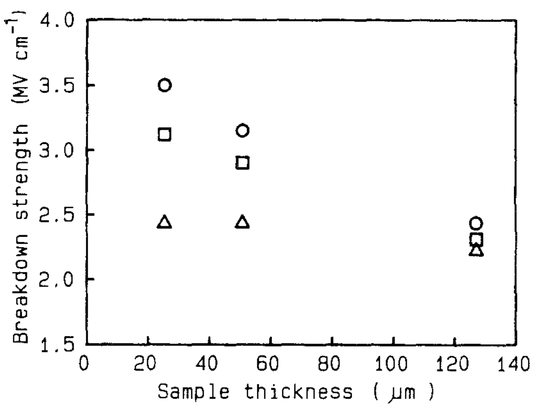

Figure 6 Sample thickness dependency of breakdown strength of PFA film at (○) 22°C, (□) 125°C, (△) 250°C

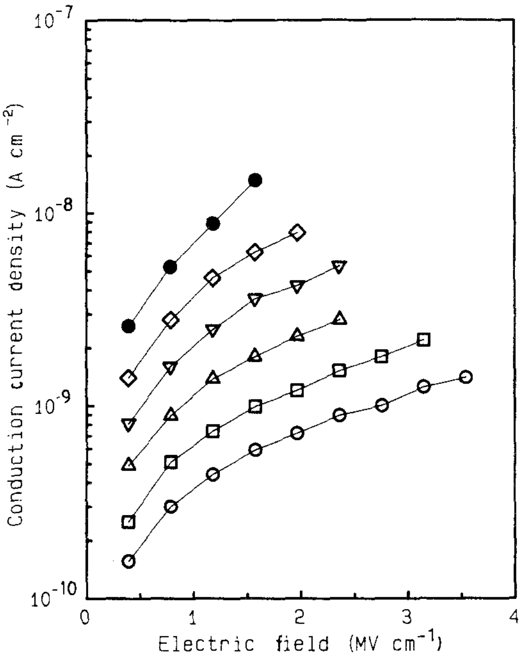

Figure 7 *J–F* curves of PFA film at (○) 22°C, (□) 50°C, (△) 100°C, (▽) 150°C, (◇) 200°C, (●) 250°C.

4. Discussion

The presence of micro-voids within the film structure can greatly affect electrical properties of polymeric insulating materials. Therefore, the determination of

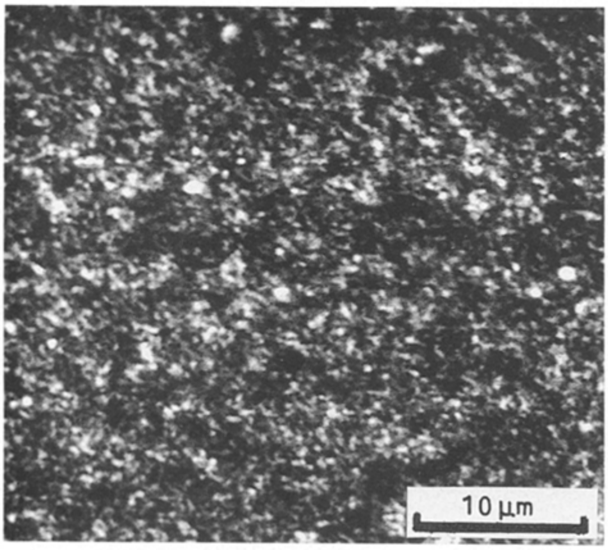

Figure 8 The microstructure of 25 µm thick PFA film at 10 µm deep from the surface using a confocal laser microscope.

micro-cavities in the dielectric material becomes crucial and facilitates explanation of changes in high-voltage properties. Such micro-voids in PFA film were identified by confocal laser microscopy which are, as shown in Fig. 8, almost submicrometre in diameter at room temperature. Small bubbles in the micro-structure of the PFA film have also been mentioned in an earlier work [12].

The characterization of the PFA film was carried out in terms of changes in their permittivity and dielectric loss as a function of frequency, temperature and voltage. The permittivity of the PFA film exhibits an independent relationship with frequency up to 100 kHz. The dielectric loss at the lower end of the frequency up to 1 kHz also remains constant. Such a null dependency of dielectric loss on frequency up to 1 kHz was also reported earlier [13]. With an increase in frequency, the dielectric loss was found to increase. At 100 kHz, for example, it had increased by about one order of magnitude of its value at 50 Hz. Bro *et al.* [13], however, has reported dielectric loss to be constant in the 10^{-4} range at frequencies up to 1 MHz.

The film suffered a slight loss in its permittivity as the temperature was increased (Fig. 3). This change in the property was permanent which indicated that chemical changes and degradation of the film could be taking place at higher temperatures [16]. The dielectric loss of the PFA film exhibited a one order increase in magnitude with a rise in temperature to 200° C. A similar increase was also observed earlier at temperatures up to 250° C and power frequency by Bro *et al.* [13], and is not consistent with Sacher's [17] data at 10 kHz. As in most cases a sudden increase in the dielectric loss is an indication of partial discharge inception [18, 19], this increase could be attributed to increased partial discharge activities in the observed micro-voids at higher temperatures and higher voltages [20].

There was no appreciable change in permittivity with applied voltages up to 1000 V, as shown in Fig. 4. However, the dielectric loss showed an abrupt increase above 400 V. As mentioned earlier, a sudden increase in dielectric loss could be associated with partial discharge activity. Such types of discharge activities were also observed on the PDA, the result (tabulated in Table II) showing sudden growth, the so-called inception level, in discharge activity at around 500 V. Therefore, it is believed that these partial discharges in micro-voids could be responsible for or at least have a major contribution to the increase in the dielectric loss of the film above the threshold value. Non-linearity of dielectric loss as a function of applied voltage has also been reported by Bartnikas for other films [21], as well as by Kreuger [18].

The breakdown strength of PFA film is almost independent of temperature up to 150° C as shown in Fig. 5. However, as temperature is further increased, a significant reduction in breakdown strength is noticed. Considering the temperature dependence of the dielectric strength, this behaviour can be roughly divided into two regions: below 150°C and above 150° C. This behaviour can also be visualized from thickness dependency results shown in Fig. 6.

In Region 1, i.e. below 150° C, the dielectric strength is almost independent of temperature. Therefore, in this region, which is considered to be a low-temperature region, the ionic conductivity is small compared to electronic conductivity at a high field. Because the ionic conductivity is too small to cause more than infinitesimal heating of the dielectric material, the electronic conductivity becomes dominant and leads to breakdown of the sample [22]. In Region 2, i.e. above 150° C, the breakdown strength decreases with an increase in temperature. There are at least two possible breakdown mechanisms which can explain the negative temperature dependence of the dielectric strength: (1) electro-mechanical breakdown theory, and (2) thermal breakdown theory. Detailed explanation of these breakdown theories can be found elsewhere [23], and, therefore, only the application is discussed below to understand the possible breakdown phenomena in the present study.

The electro-mechanical breakdown strength can be calculated using an equation given by Stark and Garton [24]

$$E_{\mathrm{M}} = \exp\left(-\frac{1}{2}\right)\left(\frac{Y}{\varepsilon}\right) \qquad (1)$$

Using the available values of Young's modulus, Y, at two temperatures [9], and measured values of permittivity, ε, from the present work, the breakdown

TABLE II Partial discharge activities in PFA film at 22° C

Voltage (V)	Partial discharge level (± 2 pC)
100	10
200	12
300	12
400	15
500	28
600	31

strength is calculated. The results are given in Table III together with the experimentally obtained values of breakdown. No agreement is found with each other, and therefore, electro-mechanical breakdown theory does not support the breakdown process taking place in PFA film at high temperatures.

The other possible mechanism, therefore, could be thermal. Thermal breakdown is usually composed of two possible extreme cases: steady state thermal breakdown which involves ignoring the time derivative; and impulse thermal breakdown by neglecting the heat conduction term [23]. Because the voltage rate in this measurement is not slow enough to obtain steady state, the first case cannot be considered for further discussion. In order to ignore the heat conduction in this breakdown process, determination of the thickness dependency on the dielectric strength of the film is one possible way [6]. As already seen from thickness dependency results shown in Fig. 6, the breakdown voltage at temperatures above 150° C does not depend on the thickness of the sample. Therefore, it is believed that joule heating does not diffuse away from the film. Therefore, heat conduction can be neglected and the thermal breakdown equation reduces to

$$C_v \frac{\partial T}{\partial t} = \sigma E^2 \tag{2}$$

It seems appropriate to conclude from these characteristics that the impulse thermal breakdown could be an operative mechanism at higher temperatures in Region 2.

In addition to the above, it is also possible that breakdown could be related to the cohesive energy density (CED) [25, 26]. Such a probability should not be ruled out and must be considered in future investigations. It is also possible that there is an existence of transition region in which electronic breakdown and thermal breakdown operate simultaneously. This phenomena can be explained in simple terms. When the applied voltage is increased, more energy is stored in the film and more dielectric loss is converted to heat, giving rise to film temperature. When the test temperature increases, heat conduction has a lesser effect on the breakdown and it might exhibit impulse-type thermal breakdown. Moreover, the conduction current also increases almost exponentially as temperature and voltage are increased, as was shown in Fig. 7. This triggers the indication of acceleration of mobility of ions and free charge carriers within a dielectric [20]. This was already suspected when an increase in dielectric loss was noticed at higher temperature but at low fields.

TABLE III Comparison of calculated electro-mechanical breakdown strength of PFA film using Stark and Garton's equation, [20] and present experimental results

Temperature (° C)	Calculated value (MV cm^{-1})	Experimental value (MV cm^{-1})
25	28.0	3.5
200	7.4	2.4

5. Conclusion

The results of present work on PFA film indicate that dielectric loss increases and dielectric strength reduces with rising temperature. Partial discharge activities primarily are responsible for an increase in the dielectric loss as the temperature and voltage are raised. The trend observed for a negative temperature dependence of breakdown strength has been divided in two regions. In Region 1, which is the low-temperature region below 150° C, it is believed that electronic conduction dominates the breakdown process. However, in Region 2, above 150° C, impulse thermal breakdown could be an operative mechanism. It is interesting to note that PFA film maintained its physical integrity and electrical properties at temperatures as high as 150° C.

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